

# FINAL REPORT

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## Comprehensive Assessment of Exposure and Lifetime Cancer Incidence Risk from Plutonium Released from the Rocky Flats Plant, 1953–1989

Part of Task 3: Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals

September 1999

*Submitted to the Colorado Department of Public Health and Environment, Disease Control and Environmental Epidemiology Division, Rocky Flats Health Studies in partial fulfillment of Contract No. 100APPRCODE 391*

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*"Setting the standard in environmental health"*



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**Part of Task 3:      Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals**

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## EXECUTIVE SUMMARY

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently contractor-operated by Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons research, development, and production complex. The RFP is located about 8 to 10 km from the cities of Arvada, Westminster, and Broomfield, Colorado, and 26 km northwest of downtown Denver, Colorado.

Through a 1989 Agreement in Principle between DOE and the State of Colorado, DOE provided the State with funding and technical support for health-related studies. The purpose of the Historical Public Exposures Studies on Rocky Flats is to estimate exposure to nearby residents from past toxic and radioactive releases from the plant. Research was performed in two phases. Phase I of the study was performed by ChemRisk (a division of McLaren/Hart Environmental Engineering). In Phase I, ChemRisk conducted an extensive investigation of past operations and releases from the RFP. *Radiological Assessments Corporation (RAC)* was awarded the contract to conduct Phase II of the study, which is an in-depth investigation of potential risks to the public from historical releases from Rocky Flats.

This report documents a comprehensive assessment of historical airborne plutonium<sup>a</sup> releases from the RFP during its operations from 1953 to 1989. This work draws heavily on the source term, atmospheric transport, cancer risk characterization, and environmental monitoring investigations previously performed during Phase I and Phase II of the Historical Public Exposures Studies on Rocky Flats.

**Methodology.** The overall objectives of this assessment are to (a) calculate concentrations of plutonium in environmental media resulting from airborne releases of plutonium at the RFP, (b) estimate incremental lifetime cancer incidence risk from inhalation of airborne plutonium to hypothetical receptors in the model domain, and (c) estimate uncertainty associated with model estimates and evaluate model accuracy. Phase I concluded that exposure was dominated by the atmospheric pathway. Therefore, calculations were limited to this pathway of exposure. Previous Phase II investigations ([Rood 1999a](#); Rood and Grogan [1999a](#), [1999b](#), [1999c](#)) focused on evaluating atmospheric transport for a single release event and estimating cancer incidence risk for hypothetical receptors residing at the location of maximum exposure and at major population centers in the model domain. The model developed in this report combines atmospheric transport simulations for multiple release events and an exposure scenario and risk coefficients to generate lifetime cancer incidence risk estimates for nearly all locations in the model domain.

We evaluated two types of uncertainty in this assessment: parameter uncertainty and model uncertainty. Parameter uncertainty addresses the uncertainty in model output given the uncertainty in model input and was used to assess model precision. Model parameters are represented by distributions of possible values. Parameter uncertainty was evaluated using Monte Carlo sampling techniques. Model uncertainty addresses the errors in the model formulation and processes. We evaluated model uncertainty (or model accuracy) by comparing model-predicted

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<sup>a</sup> In this context, the word plutonium means weapons-grade plutonium, which consists primarily of <sup>239</sup>Pu (~93.8%), <sup>240</sup>Pu (~5.8%), and <sup>241</sup>Pu (~0.36%) by weight percent. Specific activity of weapons-grade plutonium is 0.072 Ci g<sup>-1</sup>.

concentrations in environmental media with corresponding measured data. Comparing model predicted-to-observed values is often referred to as model validation.

The conceptual model segregated plutonium releases into two types: continuous and discrete. Continuous releases were characterized by annual release quantities that were assumed to be emitted to the atmosphere at a constant rate throughout the year. Discrete releases were characterized by releases that were up to 26 hours in duration. Continuous releases included routine plutonium emissions from 1953 to 1989 and suspension of contaminated soil from the 903 Area during routine meteorological conditions from 1964 to 1969. The 903 Area was a former storage site of plutonium contaminated cutting oil. Contaminated cutting oil was stored in barrels that subsequently corroded and leaked plutonium onto the soil. Discrete releases included two glove box fires that occurred in 1957 and 1969 and suspension of contaminated soil from the 903 Area during high wind events that occurred in 1968 and 1969. Annual time-integrated concentrations (*TIC*) and surface deposition were calculated at 2295 receptor nodes in the model domain. Resuspension of contaminated soil was also included in the *TIC* estimates. Soil concentrations and resuspension were dynamically modeled using the annual deposition estimates at each node, a time-dependent transit rate of plutonium into deeper soil layers, and a time-dependent resuspension factor.

Four activity-particle size ranges were considered for transport and deposition of 903 Area suspension releases: <3  $\mu\text{m}$ , 3–10  $\mu\text{m}$ , 10–15  $\mu\text{m}$ , and 15–30  $\mu\text{m}$  aerodynamic equivalent diameter. The fraction of total airborne plutonium activity attached to each size range was estimated from empirical data. The first three size ranges represent respirable particles; the last size fraction was used for deposition calculations. Suspension of activity attached to particles larger than 30- $\mu\text{m}$  aerodynamic equivalent diameter was known to have occurred. However, releases were calibrated to air monitoring data that did not detect particles greater than 30  $\mu\text{m}$  ([Weber et al. 1999](#)); consequently, release quantities were not estimated for these larger particles. These larger particles were found to be important for deposition calculations, but they were not important in terms of inhalation risk because they are not respirable.

We only considered the inhalation pathway in this evaluation. We made this decision based on Phase I results that showed soil ingestion to be a minor pathway when considering the long-term exposure to Rocky Flats effluent ([ChemRisk 1994a](#)). Unlike the individual risk reports for each release event ([Rood 1999a](#); Rood and Grogan [1999a](#), [1999b](#), [1999c](#)), inhalation of resuspended contaminated soil was included in this assessment.

Lifetime cancer incidence risk coefficients (risk per unit intake) with uncertainty for plutonium inhalation were developed by [Grogan et al. \(1999\)](#) for the four critical organs: lung, liver, bone surface, and bone marrow (leukemia). Where feasible, gender- and age-specific risk coefficients were determined. Risk coefficients were reported for three different particle size distributions having geometric mean values of 1  $\mu\text{m}$ , 5  $\mu\text{m}$ , and 10  $\mu\text{m}$  activity median aerodynamic diameter and a geometric standard deviation of 2.5 in all cases.

The *TIC* values were used to calculate plutonium intake for five receptor scenarios: laborer, homemaker, child, office worker, and student. Each receptor scenario incorporated inhalation rates that reflect the receptor's age and lifestyle. Time of exposure varied among the receptors. The laborer and homemaker scenarios assumed the receptors were present for the entire operating history of Rocky Flats (1953–1989). The office worker and student scenarios assumed the receptors were present in the model domain for the 1960s and 1970s. Uncertainty was not incorporated into the exposure scenarios; in other words, we assumed the physical attributes and

behavior of the receptors were fixed. The calculated risks are not intended to represent a population of receptors who exhibit differing behaviors.

**Model Validation.** Model-predicted concentrations were compared with measurements in four environmental media: soil, ambient air, lake and reservoir sediment, and vegetation. Concentrations in ambient air and soil were extracted directly from model output. Additional calculations were needed to evaluate concentrations in lake sediment and vegetation.

**Soil.** Model-predicted plutonium soil concentrations were compared to measured data from [Webb](#) (1996) and [Poet and Martell](#) (1972). [Webb](#) (1996) measured plutonium soil concentrations in the 0–3-cm soil layer along three, 20-km long transects extending east of the 903 Area at 60°, 90°, and 120° true bearings. Poet and Martell sampled the 0–1-cm soil layer at 28 locations surrounding the RFP. Soil concentrations were also integrated across the model domain to obtain the plutonium inventory in soil. The model-predicted inventory was compared to estimates made by other researchers who extrapolated measured soil concentrations.

Model predicted soil concentrations 2 to 10 km east of the 903 Area were generally within the range-measured values. At distances <2 km from the 903 Area, the model underpredicted concentrations while concentrations and at distances >10 km were overpredicted. Model underprediction close to the 903 Area was attributed exclusion of activity attached to >30 µm aerodynamic equivalent diameter particles. Model overprediction at distances >10 km suggested that the fraction of activity attached to fine particles (<3 µm) was probably overestimated.

Model comparisons with estimates of plutonium soil inventory suggest that while the predicted spatial distribution of activity in the model domain did not correlate exactly with the measured data, the total amount of plutonium deposited in the model domain was reasonable and within the range of values estimated by other researchers.

**Ambient Air.** Environmental measurements in ambient air near the RFP before 1970 were hampered by poor detection limits and analytical techniques that only quantified total long-lived alpha activity in air. After 1970, detection limits improved and plutonium-specific measurements were made. Therefore, comparisons with estimated concentrations were limited to post-1970 data. Model-predicted concentrations were compared with measurements at four locations: one onsite at the old RFP boundary east of the 903 Area, one at the new RFP boundary along Indiana Street, and two in the communities of Broomfield and Leyden.

Model validation for this medium applied mainly to resuspension processes because releases after 1970 were dominated, for the most part, by resuspension of contaminated soil and not routine releases from the plant operations. With the exception of several years, predicted concentrations onsite and at the new RFP boundary were generally within the range of measured values. The model underpredicted concentrations at community locations in the late 1970s and early 1980s. However, many of these measurements were below the minimum detectable concentrations and some measurements were less than background plutonium concentrations from weapons testing fallout in measured in Denver. Negative model bias during the 1970s and 1980s will not substantially underestimate inhalation exposure for persons who lived in the model domain before this time because exposures were considerably less for those two decades than in the 1950s and 1960s.

**Lake and Reservoir. Sediment.** Lake sediment, like soil, is an environmental medium in which plutonium may accumulate over time. Unlike soil, contamination tends to accumulate in bottom sediments in discrete layers that build upon the previous year layer. Thus, the temporal history of contaminant deposition can be preserved in the sediment. Sediment data were available for Standley Lake and Great Western Reservoir. Both water bodies are located east of the RFP and are susceptible to contamination from depositing airborne plumes.

Sediment core data for Standley Lake was obtained from [Hardy et al.](#) (1978). These data were used to compare the concentrations of plutonium activity in the annual sediment layer to model-predicted values for the years 1964–1976. Unfortunately, sediment data from Great Western Reservoir was complicated because substantial plutonium contamination was also carried by waterborne effluent. [Schoep and Whicker](#) (1995) estimated 87% of the *total* plutonium in the reservoir was attributed to waterborne releases. Using this percentage, we compared plutonium inventories in the reservoir attributed to airborne sources to corresponding model predictions.

The model underestimated predicted concentrations in Standley Lake sediments for the peak years of airborne deposition (1968 and 1969) and for the years that followed. We attributed this result to the model underestimating deposition in the vicinity of Standley Lake and not including transport of plutonium by soil erosion and fluvial process. Predicted plutonium inventory in Great Western Reservoir from airborne sources matched values estimated by [Schoep and Whicker](#) (1995) and [Thomas and Robertson](#) (1981) reasonably well.

**Vegetation.** Vegetation monitoring at the RFP began before the plant was operating and continued into 1953. Several samples were taken in 1955.

Predicted concentrations in vegetation generally matched the temporal trends observed in vegetation measurements taken between 1953 and 1964. The vegetation model appeared to overpredict concentrations from the 1957 fire. However, lack of specific data about vegetation sampling methods and uncertainty in the vegetation model itself made model comparisons in this medium somewhat tenuous.

**Incremental Lifetime Cancer Incidence Risks.** The spatial distribution of the total (all organs) incremental lifetime cancer incidence risk for each of the five receptor scenarios at the 5th and 95th percentiles was depicted in isopleth maps. Individual organ risks were highest for the lung, followed by the liver, bone surface, and bone marrow. The laborer had the highest risk of all scenarios because he lived in the model domain for the entire period the RFP operated and had the highest breathing rate of any of the receptors. The area of maximum risk at the 95th percentile level extended south of the plant to the intersection of Colorado 58 and Interstate 70. Maximum incremental lifetime cancer incidence risk was in the  $10^{-4}$  range (a 1 in 10,000 chance of developing cancer during a lifetime). At the 5th percentile level, the maximum cancer risk was in the  $10^{-7}$  range (a 1 in 10 million chance of developing cancer during a lifetime). A similar pattern is seen in the risk isopleths for the homemaker and child scenarios. However, cancer risks are smaller because the breathing rates for the homemaker and child were lower and exposure time for the child was shorter. Spatial distribution of risks for these receptors (laborer, homemaker, and child) were strongly influenced by the dispersion patterns from the 1957 fire.

The office worker and student scenarios showed a different spatial distribution of risk because these receptors were not present in the model domain during the 1957 fire. Maximum risks at the 95th percentile level and outside the RFP boundary for the office worker were around

$5 \times 10^{-5}$  (a 5 in 100,000 chance of developing cancer over their lifetime). Cancer risks for this scenario were dominated by releases from the 903 Area. Risks were somewhat higher for the student scenario because of an additional year of exposure to 903 Area releases (1964) and a higher breathing rate for the student compared to the office worker.

Cancer risk estimates by decade of exposure at selected locations in the model domain for the laborer scenario showed that the relative importance of each decade of exposure depended on the receptor location and the percentile level chosen. Maximum risks at the 95th percentile were dominated by exposure during the years 1953–1959 provided the receptor was located in the plume path of the 1957 fire. Otherwise, risks were dominated by releases from the 903 Area during 1964–1969. Using the laborer located about 5 km south of the RFP at Leyden as an example, the cancer risk estimates can be interpreted as follows:

- There is a 90% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was between  $2.2 \times 10^{-8}$  (5% value) and  $2.0 \times 10^{-4}$  (95% value)
- There is a 5% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was greater than  $2.0 \times 10^{-4}$
- There is also a 5% probability the model estimated risk was less than  $2.2 \times 10^{-8}$ .

Estimated cancer risks at the 95th percentile level are within the point of departure for acceptable risks established by the U.S. Environmental Protection Agency of  $10^{-6}$  to  $10^{-4}$ . However, a single grid node near the southwest corner of the RFP boundary had a 95th percentile cancer risk value of  $1.1 \times 10^{-3}$  for the laborer scenario. The spatial extent of this excursion above the U.S. Environmental Protection Agency's acceptable risk range was limited to an area no greater than 1 km<sup>2</sup>.





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## ACRONYMS

AED	aerodynamic equivalent diameter
AMAD	activity median aerodynamic diameter
CDPHE	Colorado Department of Public Health and Environment
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
GM	geometric mean
GSD	geometric standard deviation
HAP	Health Advisory Panel
HASL	(U.S. Department of Energy) Health and Safety Laboratory
HEPA	high-efficiency particulate air (filter)
LET	linear energy transfer
MDC	minimum detectable concentration
NRC	U.S. Nuclear Regulatory Commission
RAC	<i>Radiological Assessments Corporation<sup>a</sup></i>
RATCHET	Regional Atmospheric Transport Code for Hanford Emission Tracking
RBE	relative biological effectiveness
RFP	Rocky Flats Plant
TIC	time-integrated concentration
TLL $\alpha$	total long-lived alpha (activity)
TSP	total suspended particulate
UTM	universal transverse mercator

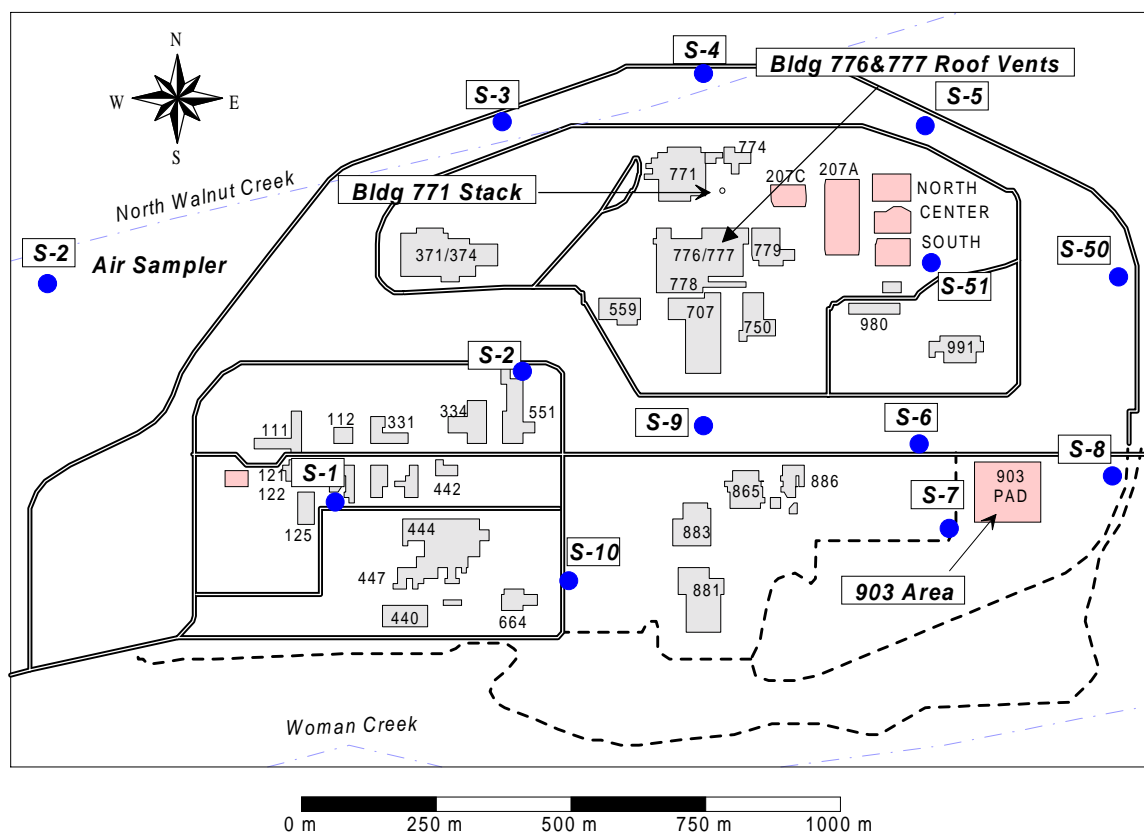
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<sup>a</sup> In 1998 *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation*. For consistency throughout the project, all reports were published by *Radiological Assessments Corporation*.



## INTRODUCTION

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently contractor-operated by Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons research, development, and production complex (Figure 1). The RFP is located on approximately 2650 ha (6500 acres) of Federal property, about 8–10 km from the cities of Arvada, Westminster, and Broomfield, Colorado, and 26 km northwest of downtown Denver, Colorado. The original 156-ha (385-acre) main production area is surrounded by a 2490-ha (6150-acre) buffer zone that now delineates the RFP boundary.



**Figure 1.** Main production area of the Rocky Flats Plant as it appeared in 1990. Originally, the buildings were identified with two-digit numbers. Later, a third digit was added. The production area, now sometimes called the industrial area, is surrounded by a security perimeter fence. Release points included in this comprehensive evaluation are the Building 771 stack, Building 776/777 roof vents, and the 903 Area.

Through a 1989 Agreement in Principle between the DOE and the State of Colorado, DOE provided the State with funding and technical support for health-related studies. The purpose of the Historical Public Exposures Studies on Rocky Flats is to estimate exposure to nearby residents from past toxic and radioactive releases from the plant. The Colorado Department of

Public Health and Environment (CDPHE) first invited a national panel of experts to help design the health studies. Because of intense public concern about Rocky Flats contamination among Denver metropolitan area residents following a Federal Bureau of Investigation raid of Rocky Flats in June 1989, the panel decided to stress public involvement and to separate the research into two major phases conducted by two different contractors to enhance accountability and credibility.

Phase I of the study was performed by ChemRisk (a division of McLaren/Hart, Environmental Engineering). In Phase I, ChemRisk conducted an extensive investigation of past operations and releases from the RFP. The Phase I effort identified the primary materials of concern, release points and events, quantities released, transport pathways, and preliminary estimates of dose and risk to offsite individuals. The conclusions from Phase I were released in a public summary document by the Health Advisory Panel (HAP) ([HAP 1993](#)); a series of task reports by ChemRisk (ChemRisk [1994a](#), [1994b](#), [1994c](#), [1994d](#), [1994e](#)); and several articles in the journal *Health Physics*.

*Radiological Assessments Corporation (RAC)* was awarded the contract to conduct Phase II of the study, which is an in-depth investigation of the potential doses and risks to the public from historical releases from Rocky Flats. Recommendations for work to be performed in Phase II are outlined in the Phase I summary document ([HAP 1993](#)).

This report documents a comprehensive assessment of historical airborne plutonium<sup>a</sup> releases from the RFP from 1953 to 1989. This work draws heavily on the source term, atmospheric transport, cancer risk characterization, and environmental monitoring investigations previously performed by RAC during Phase II of the Historical Public Exposures Studies on Rocky Flats. We provide a detailed description of the [conceptual](#) and [mathematical](#) model developed to integrate environmental transport model simulations from the various releases and propagate uncertainty to the final model predictions. [Model input requirements](#) are stated and discussed. [Predicted plutonium concentrations](#) in environmental media (air, soil, vegetation, and sediment) are presented and compared with historical and more recent measurements. Finally, [incremental lifetime cancer incidence](#) for plutonium inhalation is presented for five hypothetical receptor scenarios.

## METHODOLOGY

The overall objectives of this assessment are to (a) calculate concentrations of plutonium in environmental media resulting from airborne releases of plutonium at the RFP, (b) estimate incremental lifetime cancer incidence risk from inhalation of airborne plutonium to hypothetical receptors in the model domain, and (c) estimate uncertainty associated with model predictions and evaluate model accuracy. Previous Phase II investigations ([Rood 1999a](#); Rood and Grogan [1999a](#), [1999b](#), [1999c](#)) focused on evaluating atmospheric transport for a single release event and estimating cancer incidence risk for hypothetical receptors residing at the location of maximum exposure and at major population centers in the model domain. The model presented here combines atmospheric transport simulations for multiple release events and an exposure scenario

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<sup>a</sup> In this context, the word plutonium means weapons-grade plutonium, which consists primarily of <sup>239</sup>Pu (~93.8%), <sup>240</sup>Pu (~5.8%), and <sup>241</sup>Pu (~0.36%) by weight percent. Specific activity of weapons-grade plutonium is 0.072 Ci g<sup>-1</sup>.



and risk coefficients to generate lifetime cancer incidence risk estimates for nearly all locations in the model domain. Because of the size and complexity of the model, it was necessary to implement it on a computer using the FORTRAN programming language.

Because estimating uncertainty is an integral part of the assessment, we discuss its treatment in the model first in this report. This is followed with a description of our [conceptual model](#). The conceptual model is then described in terms of a [mathematical](#) model that calculates concentrations of plutonium in air and soil. (Concentrations of plutonium in other media were calculated in for [model validation](#) and are described in a later section.) We then extend the mathematical model to include plutonium intake estimates to a hypothetical receptor and present [lifetime cancer incidence risk estimates](#).

### **Treatment of Uncertainty**

Environmental assessment models are inherently uncertain. Uncertainty in a model arises because of (a) errors in the mathematical formations of physical processes that govern the behavior of the system (model uncertainty) and (b) errors in model input (parameter uncertainty). Model uncertainty may be evaluated by comparing model predictions to measured like quantities that are independent from the data used to develop the model. This process is often termed model validation. In some cases, models are *calibrated* to measured data, that is, model parameters are adjusted so that model predictions match the measurements.

Parameter uncertainty arises because of lack of knowledge about a parameter's true but unknown value. The uncertainty in a parameter can result from imprecise measurement of the quantity or that the parameter must be estimated because it represents a quantity that is simply impossible to measure directly. For example, the parameter may represent an average over time- and space-scales that is significantly different from measured data. To perform a parameter uncertainty analysis, probability distributions must be specified that quantitatively express the state of knowledge about each parameter considered uncertain. The distribution assigned to a parameter characterizes the degree of belief that the true, but unknown, value lies within a specified range of values ([NCRP 1996](#)). The process of establishing parameter distributions is not entirely quantitative because relevant measured data are not always available, and analysts must make judgments about the state of knowledge of the parameter.

Parameter uncertainty is propagated through the model using a variety of techniques. Analytical methods may be applied to simple models that consist of simple algebraic equations and distributions that can be defined analytically. The model presented in this report is complex enough to require the use of numerical techniques to propagate uncertainty. This study used Monte Carlo simulation combined with simple random sampling to propagate uncertainty through the model. In simple random sampling, a random value is taken from the distribution specified for each uncertain model parameter, and a single estimate of the desired endpoint is calculated. This process is repeated for a specific number of model realizations or trials. The result is an empirical approximation to the probability distribution of the model output ([NCRP 1996](#)).

Both parameter uncertainty analysis and model validation were performed for the model described in this report. Many of the input distributions were developed in other Phase II reports that documented source term, fate and transport calculations, and risk coefficients ([Rood 1999a](#); Rood and Grogan [1999a](#), [1999b](#), [1999c](#); Voillequé [1999a](#), [1999b](#), [1999c](#); [Weber et al. 1999](#);

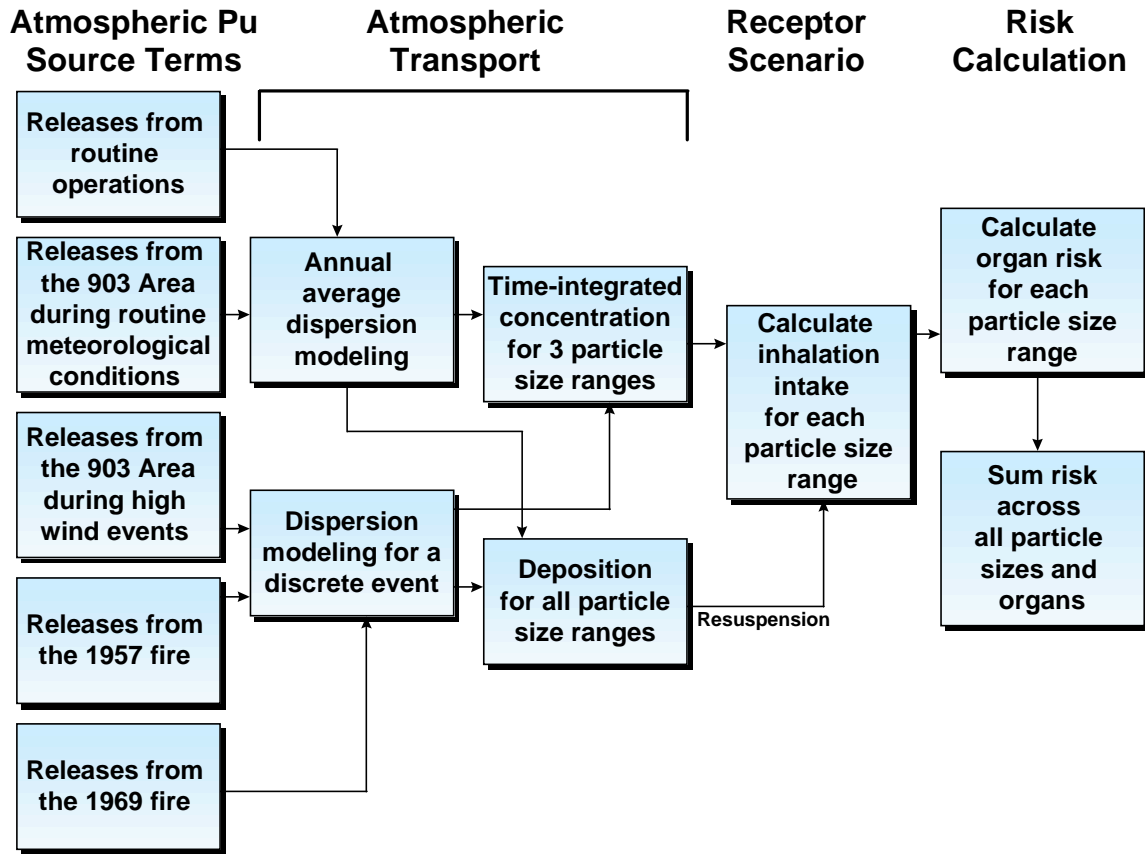
[Grogan et al. 1999](#)). The following paragraphs provide more detail about how we developed parameter distributions specific to this model.

The distributions of model output were developed from 500 model realizations. The decision to use this number was based more on computer run time and disk storage considerations than statistical considerations. While adding more realizations would result in greater confidence in the output distribution, the real question is what confidence do we have in any given percentile of the overall distribution. We defined a confidence interval around percentiles of the output distribution using a distribution-free approach developed in [Hahn and Meeker](#) (1991). The approach developed by Hahn and Meeker uses ordered statistics to define an interval where the true value of a given percentile lies at a specified level of confidence. In this way, we could define our confidence for any given percentile within the distribution. We were most interested in the tails of the distribution, because values at the top and bottom of the distribution change more with the number of model realizations; central values are more stable. The ordered statistics for the 5th and 95th percentiles for 500 model realizations is 25 and 475, respectively. That is, if the output values for 500 realizations are sorted in ascending order, the 5th percentile represents the 25th highest value; the 95th percentile represents the 475th highest value. The 95% confidence interval around the 5th percentile in terms of the ordered statistics is 15 and 35. The 95% confidence interval around the 95th percentile in terms of the ordered statistics is 465 and 485. We interpret this to mean we are 95% percent confident that 90% of the model output lies between the ordered statistics 15 and 485. The range of *values* represented by these ordered statistics will vary depending on the distribution. All model output in this report was expressed in terms of the 95% confidence interval around the 5th and 95th percentile values.

The components of uncertainty that appear in the cancer risk estimates express only uncertainty in the source terms, environmental transport, and risk coefficients. Such components of uncertainty are real in the sense they can be derived from measured quantities or inferred from historical records. Uncertainty related to the exposure scenarios was not included in this assessment. The receptor scenarios were constructed to represent hypothetical individuals living near the RFP during 1953–1989. These scenarios make it possible to examine the sensitivity of risk estimates to different assumptions about location and time of exposure to RFP releases. The scenarios might also be helpful to some individuals whose exposure history might appear similar to the scenario. We emphasize that these scenarios only represent hypothetical individuals and may not correspond to any known real individual. Physical attributes and behavior of the receptor in the scenario are not considered uncertain because this subject is not a real person, nor are they a surrogate for one or more real individuals with presumably similar behavior patterns. Rather, the calculations for this hypothetical individual are intended to show the results of possible patterns of exposure—not to approximate cancer risk that is known to have been incurred.

### Conceptual Model

A conceptual model provides an overview of the important processes, features, and assumptions that are included in a model. The conceptual model ([Figure 2](#)) is divided into four parts: source terms, atmospheric transport, exposure or intake, and cancer incidence risk. Each part of the conceptual model is discussed below.



**Figure 2.** Conceptual model for the comprehensive evaluation of exposure and risk from airborne emissions at the Rocky Flats Plant.

## Source Terms

The source term is the quantity of contaminant released to an environmental media. In this case, the contaminant is plutonium and the environmental media is air. Source terms were segregated into discrete events and those that were treated as continuous releases. Discrete events were defined as those that occurred over a relatively short period of time ( $\leq 26$  hours). They included releases from two glove box fires in 1957 and 1969 and suspension of plutonium-contaminated soil from the 903 Area during unusually high wind events in 1968 and 1969. Continuous releases included routine plutonium releases from the Building 771 stack and Building 776 roof vents and suspension of plutonium-contaminated soil from the 903 Area during routine meteorological conditions from 1964 to 1969. The development of source terms is documented in previous Phase II reports (Voillequé [1999a](#), [1999b](#); [Weber et al. 1999](#)).

Discrete 903 Area releases were modeled for 6 days in which high suspension was suspected to have occurred. Four separate particle sizes were modeled for all 903 Area releases:  $< 3 \mu\text{m}$ ,  $3\text{--}10 \mu\text{m}$ ,  $10\text{--}15 \mu\text{m}$ , and  $15\text{--}30 \mu\text{m}$  aerodynamic equivalent diameter (AED). The first three sizes are in the respirable size range, and the last was included with the others for deposition calculations. The particle sizes represent soil particles to which plutonium particles are attached.

Risk coefficients ([Grogan et al. 1999](#)) were developed for three particle size distributions that bracket the respirable particle size range used in the 903 Area simulations.

For the 1957 fire event, particles in the 1 to 10  $\mu\text{m}$  size range were modeled. For the 1969 fire event and releases from routine operations, all effluent was assumed to be in the HEPA-filtered size range, having an activity median aerodynamic diameter (AMAD) of 0.3  $\mu\text{m}$ .

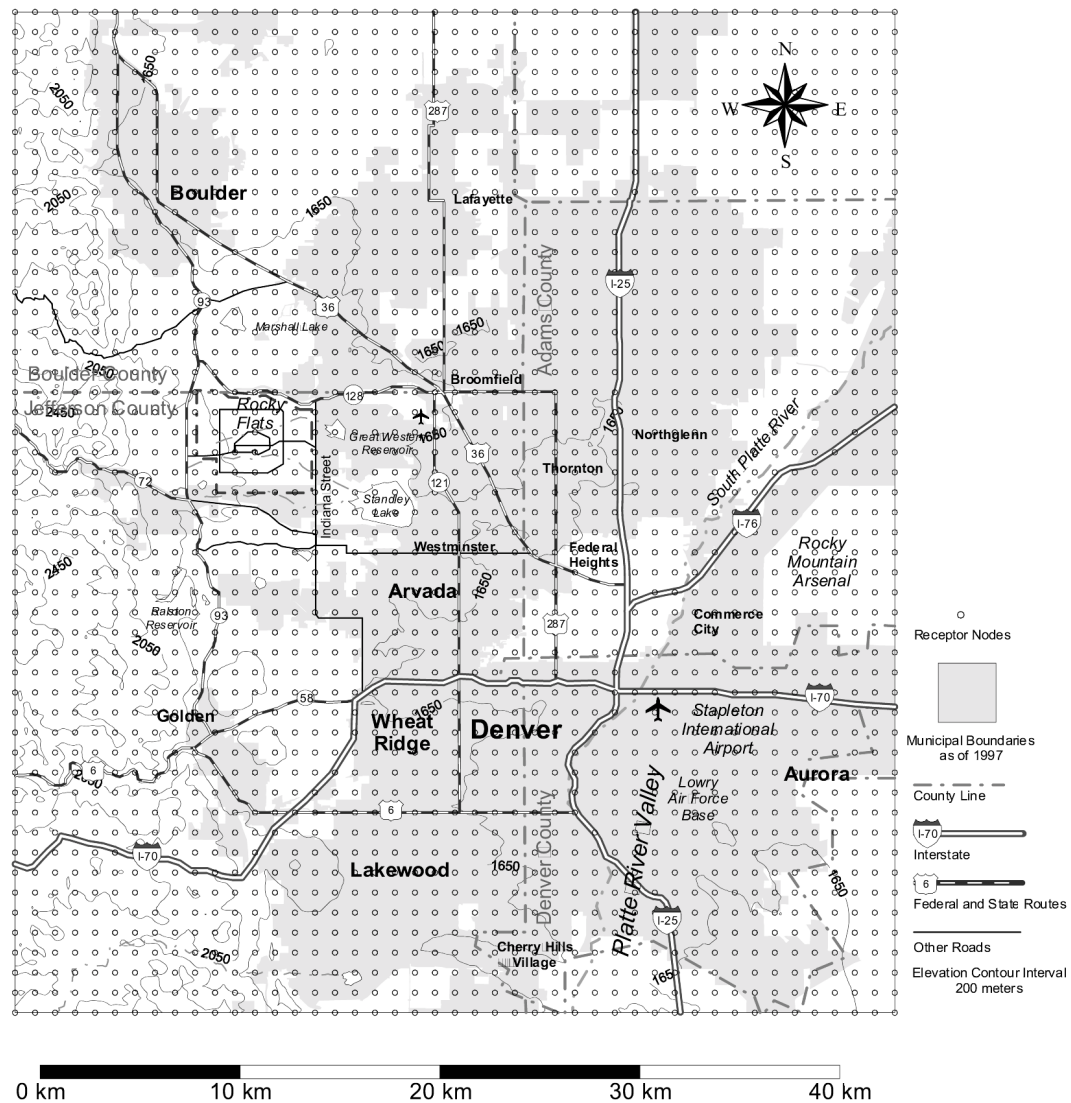
## Atmospheric Transport

Atmospheric transport modeling was performed using the Regional Atmospheric Transport Code for Hanford Environmental Tracking (RATCHET) model ([Ramsdell et al. 1994](#)). Selection of RATCHET was based on a model comparison study performed for Phase II ([Rood 1999b](#)). In this study, five models, ranging in complexity from a simple straight-line Gaussian plume model (Industrial Source Complex Short Term Version 2 [[EPA 1992](#)]) to a complex terrain model (Terrain-Responsive Atmospheric Code [[Hodgin 1991](#)]), were compared to tracer measurements taken during the 1991 Winter Validation Tracer Study ([Brown 1991](#)). The results of this evaluation indicated no one model clearly outperformed the others. However, the puff trajectory models (RATCHET, TRIAD [[Hicks et al. 1989](#)], and INPUFF2 [[Petersen and Lavdas 1986](#)]) generally had lower variability and higher correlation to observed values compared to the other models. The RATCHET model was chosen for these calculations because it incorporates spatially varying meteorological and environmental parameters. Additionally, the model includes modules that perform random sampling of the meteorological parameters, allowing for Monte Carlo analysis of uncertainty.

Atmospheric transport simulations were performed differently for discrete and continuous events. For discrete events ([Rood and Grogan 1999a, 1999b, 1999c](#)), meteorological data for the specific days of the event were available. RATCHET was run using its Monte Carlo sampling features that sampled from distributions of basic transport parameters for each Monte Carlo trial. Transport parameters that were considered stochastically included wind speed, wind direction, mixing height, precipitation, and Monin-Obukhov scaling length. Uncertainty in the source term was also included in the simulation. Output consisted of  $n$  realizations of time-integrated concentration ( $TIC$ ) and deposition at each of the 2295 computational nodes in the model domain ([Figure 3](#)).

Continuous events were modeled somewhat differently. Meteorological data from Rocky Flats for most of the assessment period were lacking. Therefore, we relied on a technique often used in prospective analysis and in retrospective analysis when historical records are lacking. This technique uses compilations of recently acquired meteorological data as a surrogate for past or future conditions and typically only applies to assessments of long-term ( $>1$  year) dispersion conditions. We employed this technique for estimating annual average plutonium concentrations from routine releases and continuous 903 Area suspension releases ([Rood 1999a; Rood and Grogan 1999b](#)) using a 5-year data set from 1989 to 1993. Uncertainty was represented using several multiplicative correction factors that accounted for uncertainty in the dispersion process, meteorology, and deposition-plume depletion.

The model domain ([Figure 3](#)) encompasses a 2200  $\text{km}^2$  area (50 km north-south  $\times$  44 km east-west). The domain extends 28 km south, 12 km west, 22 km north, and 32 km east from the RFP. Most of the Denver metropolitan area and the city of Boulder are included in the domain.



**Figure 3.** Model domain and receptor grid for Phase II Rocky Flats Dose Reconstruction Project. Note that the municipal boundaries illustrated are those as of 1997. Urban areas were much smaller during most of the period the RFP operated (1953–1989).

The domain was limited in its western extent because few receptors were present there during the RFP operations ([ChemRisk](#) 1994e) and most of the contaminant plumes traveled east and southeast of the plant.

### Exposure Modes and Risk Calculation

The only exposure mode considered was inhalation. Ingestion of plutonium in water, food, and soil are potential pathways that could have been considered in more detail. However, plutonium oxide compounds (the predominate chemical form of plutonium effluent from Rocky

Flats [[Grogan et al. 1999](#)]) are very insoluble and tend to adhere to soil, making them relatively immobile and not readily taken up by plants or accumulated in the edible portions of animal products. In contrast, the slightly soluble form of plutonium (plutonium nitrate compounds) are more readily taken up in the gastrointestinal tract. Ingestion dose conversion factors for the two forms (slightly soluble and insoluble) differ by about an order of magnitude. Phase I results ([ChemRisk 1994d](#)) indicated direct inhalation to be the dominant pathway of exposure during the early period of RFP operations (1952–1970). For the later years (1970–1989), soil ingestion and inhalation of resuspended contaminated soil became a significant component of the total dose because of the accumulation and build up of deposited plutonium in soil and smaller airborne releases. However, Phase I assumed a mixture of soluble and insoluble plutonium. Based on an oxide form of plutonium dominating at the RFP, ingestion dose is relatively small compared to inhalation. For this reason, we only considered the inhalation mode of exposure. Inhalation was modeled for two pathways: direct inhalation from airborne plumes and inhalation of resuspended activity previously deposited onto soil.

### Mathematical Model

This section describes the equations used to (a) combine *TICs* of plutonium in air and deposition onto ground surfaces from different release events, (b) calculate plutonium soil concentrations and inventory in soil, (c) calculate plutonium intake to a receptor, and (d) calculate lifetime cancer incidence risk. The equations that describe the atmospheric transport process or source terms are provided in other Phase II reports ([Rood 1999a](#); [Rood and Grogan 1999a, 1999b, 1999c](#); [Weber et al. 1999](#); [Voillequé 1999a, 1999b, 1999c](#)), which describe the processes and the methods in detail.

The model is based on a time resolution of 1 year. That is, *TICs* and risks are not reported for time periods of less than 1 year. However, *TIC* quantities may be output for any one of the discrete events by setting the continuous release source terms to zero. In this way, time resolution of less than 1 year can be obtained for any of the discrete events. Continuous source terms were described in terms of annual release quantities. Consequently, time resolution of less than 1 year is not possible for these sources.

The model was designed to propagate uncertainty through to the final output values using Monte Carlo simulation as discussed earlier. Equations are presented for a single realization (or trial) of the model.

### Time-Integrated Concentration and Risk

The *TIC* at receptor node *i*, year *j*, and particle size *k* for a single realization is

$$TIC_{i,j,k} = \sum_{m=1}^n TICd_{i,j,k,m} + \sum_{l=1}^h \chi / Q_{i,l,k} Q_{l,k,j} DF MF DeF \Delta t + TICr_{i,j,k} \quad (1)$$

where

- $TIC_{i,j,k}$  = time-integrated concentration at node *i*, for year *j* and particle size *k* (Ci-y m<sup>-3</sup>)
- n* = the number of discrete sources during year *j*
- $TICd_{i,j,k,m}$  = time-integrated concentration for discrete source *m*, at node *i*, for year *j* and particle size *k* (Ci-y m<sup>-3</sup>)



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$h$	=	number of continuous sources
$X/Q_{i,l,k}$	=	concentration divided by source term (Chi over Q) for continuous source $l$ , at node $i$ , and particle size $k$ ( $\text{y m}^{-3}$ )
$Q_{l,k,j}$	=	source term for continuous release $l$ , particle size $k$ , and year $j$ ( $\text{Ci y}^{-1}$ )
$DF$	=	dispersion correction factor (unitless)
$MF$	=	meteorological correction factor (unitless)
$DeF$	=	plume depletion and deposition correction factor (unitless)
$\Delta t$	=	time increment (1 year)
$TICr_{i,j,k}$	=	time-integrated concentration from resuspension at node $i$ , year $j$ , and particle size $k$ ( $\text{Ci-y m}^{-3}$ ).

The value of  $TICr$  was determined using a resuspension factor approach and is discussed in a later section. Values for each realization of  $TICd$  are read from previously generated files in the same order in which they were generated. This was important for the 903 Area releases because each realization was based on a unique particle size distribution. By using the same starting random number seed in the RATCHET simulations for all particle sizes, we were able to preserve the meteorological correlation that exists across all particle sizes. For example, suppose the fraction of total activity released associated with the three respirable size fractions for the  $n$ th RATCHET realization was 0.05, 0.15, and 0.05 for  $<3 \mu\text{m}$ ,  $3\text{--}10 \mu\text{m}$ , and  $10\text{--}15 \mu\text{m}$  size fraction, respectively. Input to the model for the  $n$ th realization then includes three  $TIC$  values from RATCHET, one for each size fraction. For the  $n$ th realization, sampling of the meteorological parameters in RATCHET was the same for all size fractions. In this way, the  $n$ th realization of the  $TICd$  for the first size fraction used exactly the same meteorology as the second and third size fractions.

Chi over Q ( $X/Q$ ) values were calculated using a 5-year meteorological data set spanning the years 1989 to 1993. Combined with the source term, these values were used to estimate annual average concentrations from continuous releases. Uncertainty in the dispersion estimate was accounted for by the multiplicative correction factors  $DF$ ,  $MF$ , and  $DeF$ .

Time-integrated concentrations for continuous releases were adjusted for the number of days the receptor was in the model domain. This was done by multiplying the  $TIC$  value by the ratio of the exposure time to the simulation time step (1 year). For example, if the receptor was present in the model domain 350 days per year, then the  $TIC$  for continuous sources was multiplied by the ratio of 350 d/365 d. For discrete events, the receptor was either present or not present in the model domain.

[Equation \(1\)](#) is applicable to a single year of exposure. When multiple years of exposure are considered, sampling of the three correction factors ( $DF$ ,  $MF$ ,  $DeF$ ) was done in a way to preserve correlation where it existed. The meteorological and deposition correction factors are independent from year to year ([Rood 1999a](#)), and the dispersion correction factor is correlated from year to year. That is, the dispersion correction factor applies to any long-term ( $>1$  year) estimate of air concentration while the meteorological and deposition correction factor applies to each year in the simulation. Therefore, the meteorological and deposition correction factors were sampled for each year in a realization, while the dispersion correction factor was sampled only once for each realization.

Plutonium intake by inhalation to a hypothetical receptor residing at node  $i$ , for year  $j$ , and particle size  $k$  is

$$I_{i,j,k} = TIC_{i,j,k} BR_j \quad (2)$$

where

$TIC_{i,j,k}$  = time-integrated concentration at node  $i$ , for year  $j$ , and particle size  $k$  (Ci-y m<sup>-3</sup>)  
 $BR_j$  = breathing rate for year  $j$  (m<sup>3</sup> y<sup>-1</sup>).

Note that the breathing rate may change as a function of time. This allows for the receptor to “age” during their exposure to Rocky Flats effluents. Also note that particle sizes are kept segregated throughout the calculation because risk coefficients were particle-size dependent. The total plutonium inhaled during the hypothetical receptor’s exposure period is

$$IT_{i,k} = \sum_{j=t1}^{t2} I_{i,j,k} \quad (3)$$

where

$IT_{i,k}$  = integrated inhalation intake at node  $i$ , for particle size  $k$  (Ci)  
 $t2$  = ending year of exposure (year)  
 $t1$  = beginning year of exposure (year).

The lifetime cancer incidence risk for the  $n$ th organ of interest is

$$R_{i,n} = \sum_{k=1}^3 I_{i,k} RC_{k,n} \quad (4)$$

where

$R_{i,n}$  = incremental lifetime cancer incidence risk for organ  $n$  at node  $i$   
 $RC_{k,n}$  = risk coefficient for particle size  $k$  and organ  $n$ .

The total incremental lifetime cancer incidence risk from all organs is given by

$$Rt_i = \sum_{n=1}^4 R_{i,n} \quad (5)$$

where

$Rt_i$  = total incremental lifetime cancer incidence risk at node  $i$ .

In the formulation presented, the risk coefficients are sampled once for each realization in the Monte Carlo simulation. Age- and gender-specific risk coefficients were developed for Phase II and are summarized in a later section of this report. Age groups were separated into those younger than 20 and those 20 years and older. The risk coefficients used in [Equation \(4\)](#) were based on the age of the receptor at the start of their exposure. We acknowledge that this introduces some inaccuracies when the receptor matures beyond the age of 20 during their exposure. We have designed the scenarios presented in this report so that this condition does not occur. Nevertheless, this inaccuracy is not expected to have a significant impact on the overall distribution of risks for a receptor who does mature beyond the age of 20 during their exposure.



## Plutonium Concentration and Inventory in Soil

We used soil concentration estimates to calculate resuspension of plutonium-contaminated soil, compare model-predicted values with measured soil and sediment concentrations, and compute plutonium inventories in the model domain. The surface deposition at receptor node  $i$  and year  $j$  for a single realization is

$$SD_{i,j} = \sum_{m=1}^n SDd_{i,j,m} + \sum_{l=1}^h \chi / Qd_{i,l} Q_{l,j} DF MF DeF \Delta t \quad (6)$$

where

$SD_{i,j}$	=	surface deposition at node $i$ and year $j$ ( $\text{Ci m}^{-2}$ )
$n$	=	the number of discrete sources during year $j$
$SDd_{i,j,m}$	=	surface deposition for discrete source $m$ , at node $i$ , and year $j$ ( $\text{Ci m}^{-2}$ )
$h$	=	number of continuous sources
$X/Qd_{i,l}$	=	surface deposition divided by source term for continuous source $l$ at node $i$ ( $\text{y m}^{-2}$ )
$Q_{l,j}$	=	source term for continuous release $l$ and year $j$ ( $\text{Ci y}^{-1}$ )
$DF$	=	dispersion correction factor (unitless)
$MF$	=	meteorological correction factor (unitless)
$DeF$	=	plume depletion and deposition correction factor (unitless)
$\Delta t$	=	time increment (1 year).

For surface deposition calculations, particle size was not distinguished, and surface concentrations represent all particle sizes  $<30\text{-}\mu\text{m}$  AED. Larger particles were known to have been suspended from the 903 Area during high wind events, but they were not included in the release estimates because air sampler data that were used to calibrate the 903 Area release model failed to detect most particles  $>30\text{ }\mu\text{m}$  (Weber et al. 1999). These larger particles would tend to deposit close to the source (1–2 km) and are not respirable. Therefore, near the 903 Area we expect the model to underpredict soil concentrations. For 903 Area baseline releases, we performed the deposition calculations using the source term for all particle sizes ( $<30\text{ }\mu\text{m}$ ) and a gravitational settling velocity based on an activity-weighted mean particle size. The activity-weighted mean particle size (Table 1) is the sum of the midpoint of the particle size range times the fraction of activity in that range. Particle size ranges and the fraction of activity in each size range were obtained from Langer (1986) and are consistent with those used to develop the 903 Area source term (Weber et al. 1999).

The total amount of plutonium deposited in soil at end of an exposure period is given by

$$SDT_i = \sum_{j=1}^n SD_{i,j} \quad (7)$$

where

$SDT_{i,j}$	=	total surface deposition at node $i$ for $n$ years of deposition ( $\text{Ci m}^{-2}$ )
$n$	=	the number of years from the start of RFP operation to the end of exposure.

**Table 1. Activity-Weighted Mean Particle Size used for Baseline 903 Area Deposition Calculations (based on [Langer 1986](#))**

Particle size range AED ( $\mu\text{m}$ )	Midpoint of particle size ( $\mu\text{m}$ )	Fraction of airborne activity in size range
<3	1.5	0.035
3–10 <sup>a</sup>	5.5	0.0775
10–15 <sup>a</sup>	12.5	0.0775
15–30	22.5	0.81
Activity-weighted mean particle size ( $\mu\text{m}$ )		20.

<sup>a</sup> Langer reports only the 3–15  $\mu\text{m}$  size range. We assumed particles were equally distributed between 3–10  $\mu\text{m}$  and 10–15  $\mu\text{m}$ .

The model begins the simulation at the start of RFP operations (1953). This allows for all RFP releases to accumulate in the soil up to the time of exposure and calculation of resuspended plutonium in air. Provisions were made within the code to output the deposition for a single year. We used this option to estimate plutonium concentrations in annual accumulations of sediment on the bottom of nearby lakes and reservoirs.

Annual surface deposition was converted to soil concentrations using

$$SC_i = \frac{SDT_i f}{\rho T} \quad (8)$$

where

- $SC_i$  = soil concentration at node  $i$  ( $\text{Ci g}^{-1}$ )
- $f$  = fraction of plutonium inventory in soil layer of thickness  $T$
- $\rho$  = average soil density in soil layer of thickness  $T$  ( $\text{g m}^{-3}$ )
- $T$  = thickness of soil layer of interest (m).

Values for  $f$ ,  $\rho$ , and  $T$  were determined from data presented in [Webb](#) (1996) and are discussed in the section on model inputs later in this report.

The plutonium soil inventory was calculated by integrating  $SDT_i$  across the model domain.

$$I = \int_0^x \int_0^y SDT dx dy \quad (9)$$

where

- $x$  = east-west length of the model domain (44 km)
- $y$  = east-west length of the model domain (50 km).

Soil inventory is approximated for each realization by summing the  $SDT_i$  values for all nodes. The inventory estimates are then sorted in ascending order, keeping track of the realization that is associated with each inventory estimate. The code then outputs the surface deposition values associated with the sorted inventory estimates in terms of percentiles in 5% increments and the 2.5% and 97.5% values. The surface deposition values were then numerically integrated using the Surfer™ software ([Golden Software](#) 1996) to obtain an inventory estimate.

## Resuspension

Exposure to airborne plutonium via inhalation of resuspended contamination was not included in the risk reports for each individual release event ([Rood 1999a](#); Rood and Grogan [1999a](#), [1999b](#), [1999c](#)). This pathway was omitted in the individual release event evaluations because the source for resuspension (that is, the concentration of plutonium in soil) had contributions from all RFP releases. (However, it was shown that discrete 903 Area releases were responsible for most of the offsite contamination.) We included the inhalation of resuspended contamination pathway in this comprehensive evaluation.

It is important to distinguish between suspension and resuspension. Suspension describes the wind-driven movement of particles from an initially contaminated soil surface. Resuspension describes wind-driven movement of particles previously deposited from an airborne source. Releases during the 1960s from the 903 Area were classified as suspension events. Redistribution of activity deposited on the soil from 903 Area suspension releases were classified as resuspension. Many factors affect resuspension, including the particle size of the deposited material; length of time from the initial deposit (i.e. weathering); vegetative cover; and soil disturbance by animal and human activity.

Several important resuspension studies were performed at the RFP in the 1970s and 1980s. [Sehmel and Orgill](#) (1972) developed a plutonium resuspension model for the field east of the 903 Area based on saltation of bare soil surfaces. They estimated resuspension increases as a function of wind speed raised to the 2.1 power. Other studies by Sehmel included a 1973 field experiment in which airborne plutonium concentrations as a function of respirable particle size were measured ([Sehmel and Lloyd](#) 1976) and an evaluation of plutonium attached to larger than respirable particles ([Sehmel](#) 1976). [Krey et al.](#) (1974) measured plutonium in air east of the 903 Area over a 3-week period, and it reported that the total mass and plutonium concentrations in air were inversely related to precipitation and soil moisture. In addition, the “hot particle” problem and sampling bias were believed to render those short-term air measurements at the RFP uncertain and possibly nonrepresentative of annual average conditions.

In one of the more exhaustive studies, [Langer](#) (1986) measured mass loading, plutonium concentration in air, and plutonium activity concentration of resuspended dust over a 2-year period (1982–1984) at three heights (1, 3, and 10 m) and three size fractions (<3  $\mu\text{m}$ , 3–15  $\mu\text{m}$ , and >15  $\mu\text{m}$  AED). This work provided the basis for estimating activity particle size distributions used to model Phase II 903 Area suspension releases.

Monthly average mass loading during the period Langer measured ranged from 35 to 67  $\mu\text{g m}^{-3}$  at the 1-m level, and most of the activity was associated with particles >15  $\mu\text{m}$  AED. In a later study, Langer concluded that resuspension from bare soil appears to be minimal, while resuspension from grass appears to be the dominant process ([Langer](#) 1991). Over 90% of the resuspended plutonium was associated with soil and grass litter >3  $\mu\text{m}$  diameter AED. His conclusions were based on measurements performed using portable wind tunnels and acoustic particle counters. These observations seem to contradict the earlier work of [Sehmel and Orgill](#) (1972), and/or suggested that conditions at the RFP east field over time have changed. At the time of Sehmel’s measurements, more bare soil was postulated to have been exposed, resulting in greater susceptibility to soil erosion processes.

Other processes that affect resuspension are redistribution and loss of plutonium in the surface soil. These processes include soil erosion ([Webb](#) 1992, [Webb et al.](#) 1993); colloidal

movement ([Bates et al. 1992](#)); biotic perturbation ([Litaor et al. 1994](#); [Winsor and Whicker 1982](#)); and soil cracking ([Higley 1994](#)). In summary, these processes are not well understood and are currently an area of research at the RFP.

Numerous models exist for resuspension, ranging from simple empirical models to more elaborate models. The more elaborate models require extensive field data and a thorough understanding of the controlling processes. We had neither of these at the time of this study, and as previously mentioned, they are an area of current research. Therefore, our only recourse was to use one of the simple empirical models that can be implemented using a few parameters that are readily available.

Two of the most common empirical models are the resuspension factor and mass loading models ([Smith et al. 1982](#)). The resuspension factor is derived from measurements of activity concentration in air (in curies per cubic meter) and corresponding measurements of soil surface activity concentrations (in curies per square meter) in an area directly below the air measurement site. The resuspension factor is given by the ratio

$$Rf = \frac{Ca}{Cs} \quad (10)$$

where

- $Rf$  = resuspension factor ( $\text{m}^{-1}$ )
- $Ca$  = activity concentration in air ( $\text{Ci m}^{-3}$ )
- $Cs$  = surface activity concentration in soil ( $\text{Ci m}^{-2}$ ).

Resuspension factors at Rocky Flats were estimated by [Langer](#) (1991) to range from  $10^{-13}$  to  $10^{-10} \text{ m}^{-1}$ . Total resuspension flux from the 903 Area east field was estimated to be around  $200 \mu\text{Ci y}^{-1}$  (as measured in the late 1980s). In an earlier study, [Sehmel and Orgill](#) (1972) reported the resuspension factor ranged from  $10^{-9}$  to  $10^{-5} \text{ m}^{-1}$ . These measurements were made during site remediation activities, when the field east of the 903 Area was free of vegetation. Volchok reports in [Linsley](#) (1978) that resuspension factors at the RFP ranged from  $10^{-9}$  to  $10^{-6}$ . The difference between the two estimates was attributed to the soil sampling depth. The lower resuspension factor ( $10^{-9} \text{ m}^{-1}$ ) was based on a soil sampling depth of 20 cm while the higher measurement ( $10^{-6} \text{ m}^{-1}$ ) was based on a soil sampling depth of  $\approx 0.2 \text{ mm}$ .

Weathering and migration of surface deposits into deeper soil tend to decrease resuspension rates over time ([Stewart 1967](#)). Several empirical relationships have been developed to account for these processes. [Anspaugh et al.](#) (1975) developed an empirical relationship describing the decrease in resuspension over time based on measurements at the Nevada Test Site. The time-dependent resuspension factor was given by

$$Rf(t) = Rf_o e^{-\lambda \sqrt{t}} + Rf \quad (11)$$

where

- $Rf_o$  = initial resuspension factor ( $1.0 \times 10^{-4} \text{ m}^{-1}$ )
- $\lambda$  =  $0.15 \text{ d}^{-1}$
- $Rf$  = long-term resuspension factor ( $1.0 \times 10^{-9} \text{ m}^{-1}$ ).

Based on this work and others, the U.S. Nuclear Regulatory Commission (NRC) developed several “generic” equations for describing the decrease in the resuspension rate over time. The

NRC equation used in the *Reactor Safety Study—An Assessment of Accidents and Risks in Commercial Nuclear Power Plants* (NRC 1975) is

$$Rf = A e^{-0.6769t} + B \quad (12)$$

where

- $A$  = initial resuspension factor ( $m^{-1}$ )  
 $t$  = time measured in years  
 $B$  = long-term resuspension factor ( $m^{-1}$ ).

The values suggested for  $A$  and  $B$  by the NRC are  $10^{-6}$  and  $10^{-9}$ , respectively. Site-specific data suggest the initial resuspension factor ( $A$ ) is in the range measured by [Sehmel and Orgill](#) (1972), which measured resuspension shortly after paving of the 903 Area. Later measurements made in the late 1980s by [Langer](#) (1991) suggest the long-term resuspension rate is lower than the generic value ( $B$ ) suggested by the NRC.

The mass loading factor is derived in a similar manner to the resuspension factor, but it relies on dust loading measurements and was the approach used by ChemRisk in Phase I to estimate resuspension. The air concentration from resuspension is estimated using

$$C_a = ML C_s \quad (13)$$

where

- $C_a$  = airborne concentration from resuspension  
 $ML$  = mass loading factor ( $g\ m^{-3}$ )  
 $C_s$  = surface soil concentration ( $Ci\ g^{-1}$ ).

Mass loading in the vicinity of Rocky Flats has been measured by the CDPHE. [Hodgin](#) (1998) provided a review of these data for 1995, 1996, and 1997. The annual average geometric mean (GM) total suspended particulates (TSPs) was  $37\ \mu g\ m^{-3}$  at the east end of the industrial area and  $27\ \mu g\ m^{-3}$  in the interior of the east Buffer Zone. Geometric mean TSP concentrations around the perimeter of the RFP ranged from a high of  $39.8\ \mu g\ m^{-3}$  at the west perimeter to a low of  $25.6\ \mu g\ m^{-3}$  at the northern perimeter of the site. Raw data from the perimeter monitoring stations were also provided by personnel at CDPHE for the years 1992–1998. The GM of the six annual average TSP concentrations was  $35\ \mu g\ m^{-3}$ , with a GSD of 1.25. ChemRisk reported ([ChemRisk](#) 1994c) the GM TSP annual average concentration at Rocky Flats was  $60\ \mu g\ m^{-3}$  for 1980, 1983, and 1984. This value was obtained verbally from CDPHE personnel<sup>b</sup>, and records of this information were not obtained. We queried CDPHE about this value, and they referred us to the later measurements that were considerably lower than those used by ChemRisk in Phase I. A mean of  $35\ \mu g\ m^{-3}$  appears more reasonable than  $60\ \mu g\ m^{-3}$  based on other measurements near Rocky Flats and elsewhere. According to the U.S. Environmental Protection Agency (EPA) ([EPA](#) 1990), the annual mean TSP concentration at 30 nonurban sites ranged from 5 to  $50\ \mu g\ m^{-3}$ . [Whicker and Schultz](#) (1982) gives an average dust loading in nonurban locations of about  $40\ \mu g\ m^{-3}$ . For urban locations, [Gilbert et al.](#) (1983) reports TSP concentrations range from 33 to  $250\ \mu g\ m^{-3}$ .

<sup>b</sup> Personal communication with N.D. Chick, Colorado Department of Health, Air Pollution Division, June 19, 1992.

While the mass loading approach is attractive because the parameters are easily obtained, it suffers from several problems. First, there it is not always a linear relationship between surface soil plutonium concentrations and concentrations of plutonium on suspended dust. Second, weathering tends to make activity less susceptible to resuspension and the model fails to account for these processes that are well documented in other studies.

Both models (resuspension factor and mass loading) assume the air concentration is related to the soil concentration directly below. These models fail to account for upwind sources of activity and dilution from uncontaminated dust. Incorporating these processes into the model would take a considerable effort that we believe is not warranted for two reasons. First, calculations have shown that exposure during the early years of RFP operations (1953–1970) was substantially higher than exposure during the later years. Second, while resuspension is believed to be a significant component of exposure during the later years (1970–1989), measured air concentrations during this time are relatively low, resulting in overall low exposure compared to previous years. For example, monthly average respirable plutonium concentrations in the 903 Area east field measured by Langer from 1982 to 1984 ranged from  $<0.001$  to  $0.011$  fCi  $\text{m}^{-3}$ . Background plutonium concentrations in air from fallout sources at this time were about  $0.001$  fCi  $\text{m}^{-3}$ .

Based on the above discussion, we implemented the time-dependent resuspension factor approach for estimating soil resuspension from RFP releases. We used the NRC formulation of the time-dependent resuspension model ([Equation 12](#)) to predict resuspension, substituting site-specific data for the resuspension factor. We found no compelling argument for using Anspaugh's model over that of the NRC. Anspaugh's model was specific to the Nevada Test Site, while the NRC model was believed to be more generic. Because we do not have detailed site-specific data from which to develop our own model, we chose the generic model as being more applicable to wider variety of environments. [Equation \(12\)](#) applies to a single release at  $t = 0$ . The time-dependent resuspension from transient deposition over time is found using the convolution integral. The time-integrated concentration from resuspension as a function of time is given by

$$TICr(t)_{i,k} = f_k \int_0^t SD(\tau)_i Rf(t - \tau) f(t - \tau) d\tau \quad (14)$$

where

- $TICr(t)_{i,k}$  = time-integrated concentration at node  $i$ , and particle size  $k$  at time  $t$  (Ci-y  $\text{m}^{-3}$ )
- $f_k$  = fraction of the airborne plutonium activity associated with particle size  $k$
- $SD(\tau)_i$  = surface deposition at time  $\tau$  and node  $i$  (Ci  $\text{m}^{-2}$ )
- $Rf(t - \tau)$  = time-dependent resuspension factor at time,  $t - \tau$  ( $\text{m}^{-1}$ )
- $f(t - \tau)$  = fraction of the total plutonium inventory in the resuspension layer (0–3 cm) at time  $t - \tau$ .

## Model Inputs

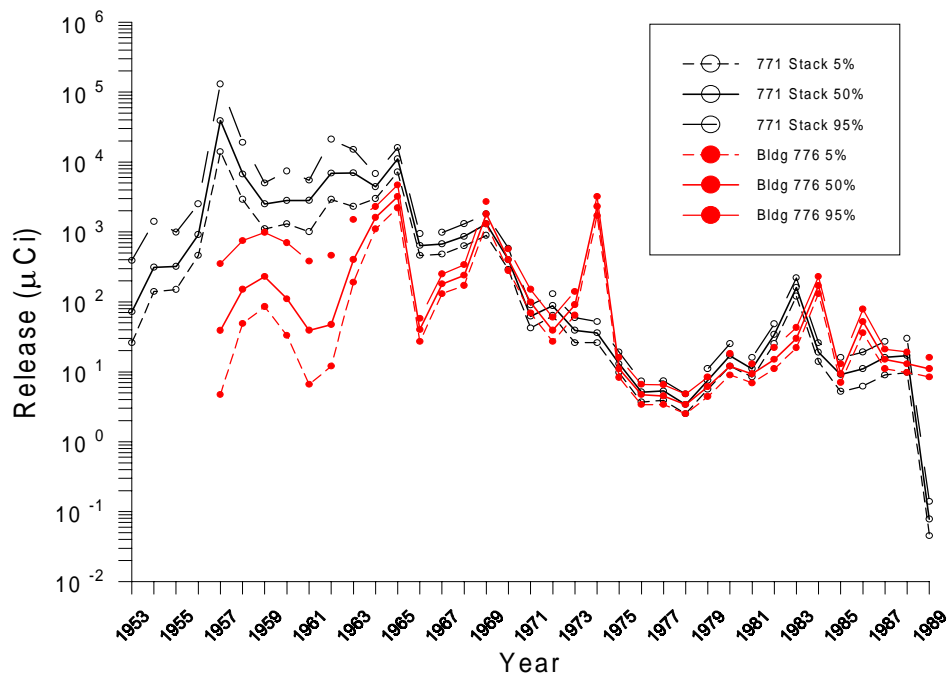
Most model input was taken from previously published Phase II reports documenting source terms, dispersion factors, *TICs*, exposure scenarios, and risk coefficients. Other input developed specifically for this model included plutonium depth distribution in soil, soil density as a function of depth, and resuspension factors. This section summarizes input requirements for the model and elaborates on parameters that were developed specifically for this model.

## Source Terms

Source terms for routine releases and baseline 903 Area releases are required model input. Quantities of plutonium routinely released from the Building 771 stack and Building 776/777 roof vents were evaluated in [Voillequé \(1999a\)](#). Median release estimates and the 5th and 95th percentile values for Building 771 stack and Building 776/777 roof vents are illustrated in Figure 4. Before 1963, releases were dominated by the Building 771 stack. The highest releases were estimated to be in 1957, the year of the fire in Building 771. Release estimates for that year ranged from 14,000  $\mu\text{Ci}$  (5<sup>th</sup> percentile) to 130,000  $\mu\text{Ci}$  (95<sup>th</sup> percentile) and were due primarily to the extended cleanup work that followed the accident. Releases as a direct result of the two major fires in 1957 and 1969 are excluded from these routine release estimates. Uncertainty in release estimates varies from year to year and is greatest during the early years of plant operation.

Effluent was passed through high-efficiency particulate air (HEPA) filters before discharge to the atmosphere. The median particle size for HEPA-filtered effluent was reported to be 0.3  $\mu\text{m}$  ([Voillequé 1999a](#)). Particle sizes from routine releases were not expected to exceed 1.0  $\mu\text{m}$ .

Suspension releases of plutonium-contaminated soil from the 903 Area for the years 1964–1969 are evaluated in [Weber et al. \(1999\)](#). The 903 Area, located in the eastern part of the main production area of the RFP, served as a waste storage area for barrels containing plutonium-laden cutting oil and degreasing agents during the late 1950s and 1960s. The barrels were stored outside on a grassy area that became known as the 903 Area. Four thousand seven hundred and



**Figure 4.** Estimates of routine releases of plutonium from the Building 771 stack and Building 776/777 roof vents. Estimates were taken from [Voillequé \(1999a\)](#) and do not include releases from the major fires in 1957 and 1969.



twenty-nine barrels were reported to have been stored at the 903 Area. By 1964, there was evidence of large-scale corrosion and subsequent leakage of barrel contents onto the soil. In 1967, efforts were made to remove the barrels from the 903 Area, repackage the contents, and ship the waste offsite. Drum removal was completed in 1968 and 1969, and the now empty area was cleared of vegetation, graded, and paved.

All 903 Area releases were calculated from a wind speed-dependent suspension model. The suspension model was calibrated to air monitoring data taken at the S-8 sampler located about 100 m east of the 903 Area. The baseline releases represent those releases that occurred during routine meteorological conditions. Analysis of S-8 sampler data and meteorological data from the Jefferson County Airport indicated high releases from the 903 Area were strongly correlated with high wind days. Releases during high wind days were treated separately as six discrete events. Originally, 24 discrete events were evaluated, but it was found that about 90% of the activity released from discrete events was attributed to only 6 of the 25 days. To reduce computational time, activity released during the 19 remaining discrete days was added to baseline releases for their respective years (Table 2). Release quantities were segregated into four particle-size classes representing <3 µm, 3–10 µm, 10–15 µm, and >15 µm. All particle sizes are reported in terms of their AED. The AED is given by

$$AED = d_p \sqrt{\frac{\rho_s}{\rho_u}} \quad (15)$$

where

- $AED$  = aerodynamic equivalent diameter (µm)  
 $d_p$  = physical diameter (µm)  
 $\rho_s$  = particle density (g cm<sup>-3</sup>)  
 $\rho_u$  = unit particle density (1.0 g cm<sup>-3</sup>).

The fraction of activity associated with each size fraction was based on measurements made by Langer (1986). Langer (1986) only reported three size fraction: <3 µm, 3–15 µm, and >15 µm AED. We split the 3–15 µm size fractions into 3–10 µm and 10–15 µm size fractions and

**Table 2. Calibrated 903 Area Release Estimates of Plutonium Attached to < 30 µm Soil Particles for the Baseline Releases (1964–1969) and Baseline plus Discrete Event Remainder**

Year	Baseline release (Ci)			Baseline plus remainder <sup>a</sup> (Ci)		
	50th %	95th %	5th %	50th %	95th %	5th %
1964	$6.6 \times 10^{-3}$	$3.8 \times 10^{-2}$	$8.1 \times 10^{-4}$	$6.6 \times 10^{-3}$	$3.8 \times 10^{-2}$	$8.1 \times 10^{-4}$
1965	$1.3 \times 10^{-2}$	$1.0 \times 10^{-1}$	$9.4 \times 10^{-4}$	$1.3 \times 10^{-2}$	$1.0 \times 10^{-1}$	$9.4 \times 10^{-4}$
1966	$1.4 \times 10^{-2}$	$1.1 \times 10^{-1}$	$1.3 \times 10^{-3}$	$1.4 \times 10^{-2}$	$1.1 \times 10^{-1}$	$1.3 \times 10^{-3}$
1967	$2.6 \times 10^{-2}$	$1.6 \times 10^{-1}$	$3.0 \times 10^{-3}$	$2.6 \times 10^{-2}$	$1.6 \times 10^{-1}$	$3.0 \times 10^{-3}$
1968	$5.5 \times 10^{-2}$	$3.0 \times 10^1$	$1.1 \times 10^{-2}$	$3.0 \times 10^{-1}$	$1.6 \times 10^0$	$1.2 \times 10^{-1}$
1969	$6.4 \times 10^{-2}$	$3.4 \times 10^{-1}$	$1.3 \times 10^{-2}$	$1.5 \times 10^{-1}$	$9.3 \times 10^{-1}$	$5.3 \times 10^{-2}$

<sup>a</sup> The remainder is the activity released from the 25 highest S-8 sampler days that were not treated as discrete events. This activity was added to the baseline releases for their respective year of occurrence.



assumed equal weighting of activity within each fraction. The fraction of activity associated with each size fraction was then 3.5%, 7.75%, 7.75%, and 81% for  $<3\ \mu\text{m}$ ,  $3\text{--}10\ \mu\text{m}$ ,  $10\text{--}15\ \mu\text{m}$ , and  $>15\ \mu\text{m}$ , respectively.

### Dispersion Factors

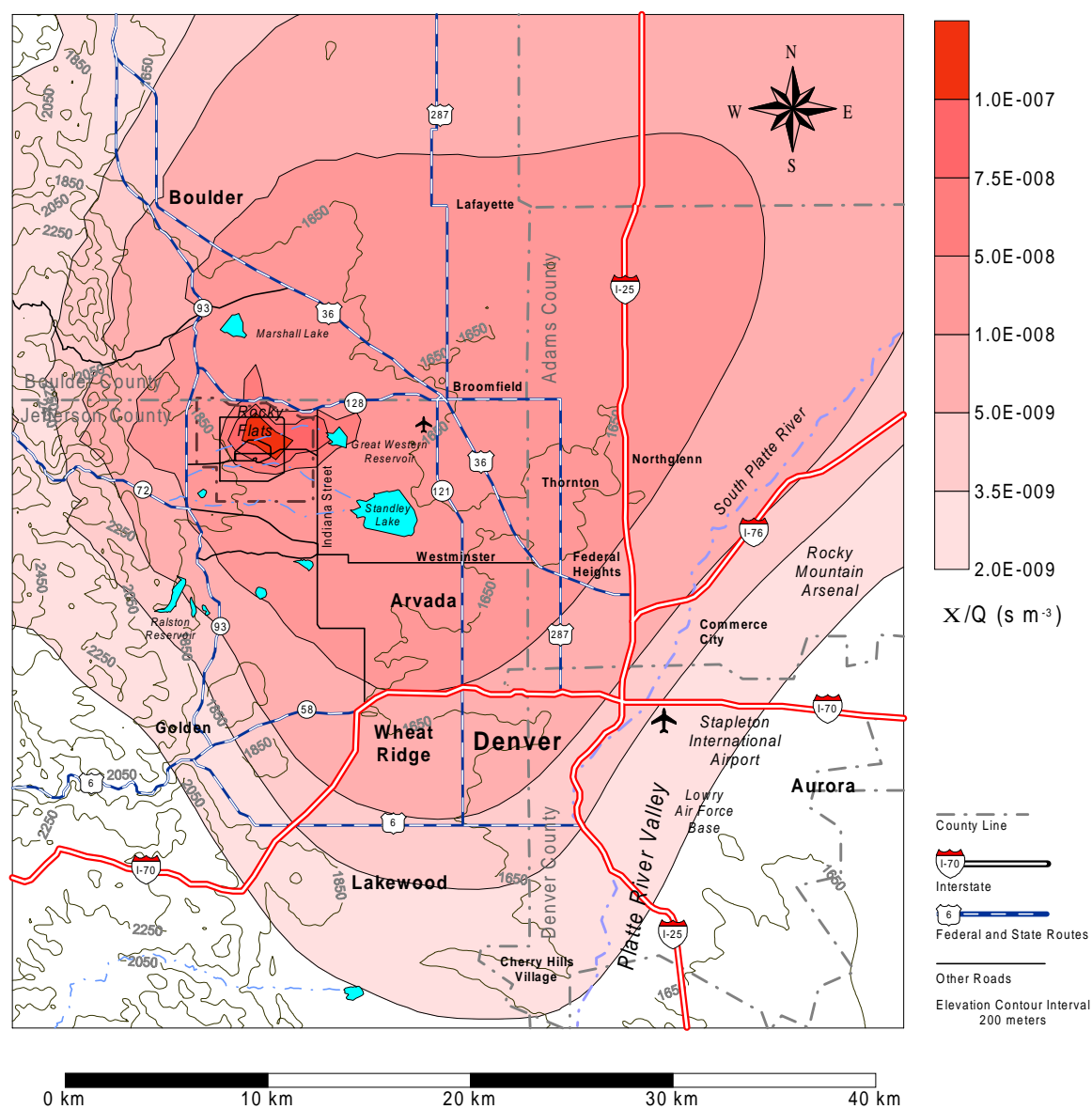
Dispersion factors or  $X/Q$  values (in seconds per cubic meter) are used with source terms for continuous events to estimate atmospheric concentrations in the model domain. Dispersion factors were calculated using the RATCHET code and a 5-year meteorological data set taken from 1989 to 1993 at the RFP, along with concurrent data taken at the Denver Stapleton International Airport.

Dispersion factors were estimated for releases from the 44-m Building 771 stack, Building 776 roof vents (Rood 1999a), and 903 Area baseline releases (Rood and Grogan 1999b). For 903 Area baseline releases, dispersion factors for three separate particle sizes were calculated. Dispersion factors at ground level for the Building 771 stack, Building 776 roof vents, and 903 Area are illustrated in Figures 5 through 7. Note that the pattern of dispersion is different for the Building 771 stack compared to the 903 Area and Building 776 because of the higher release height for stack releases.

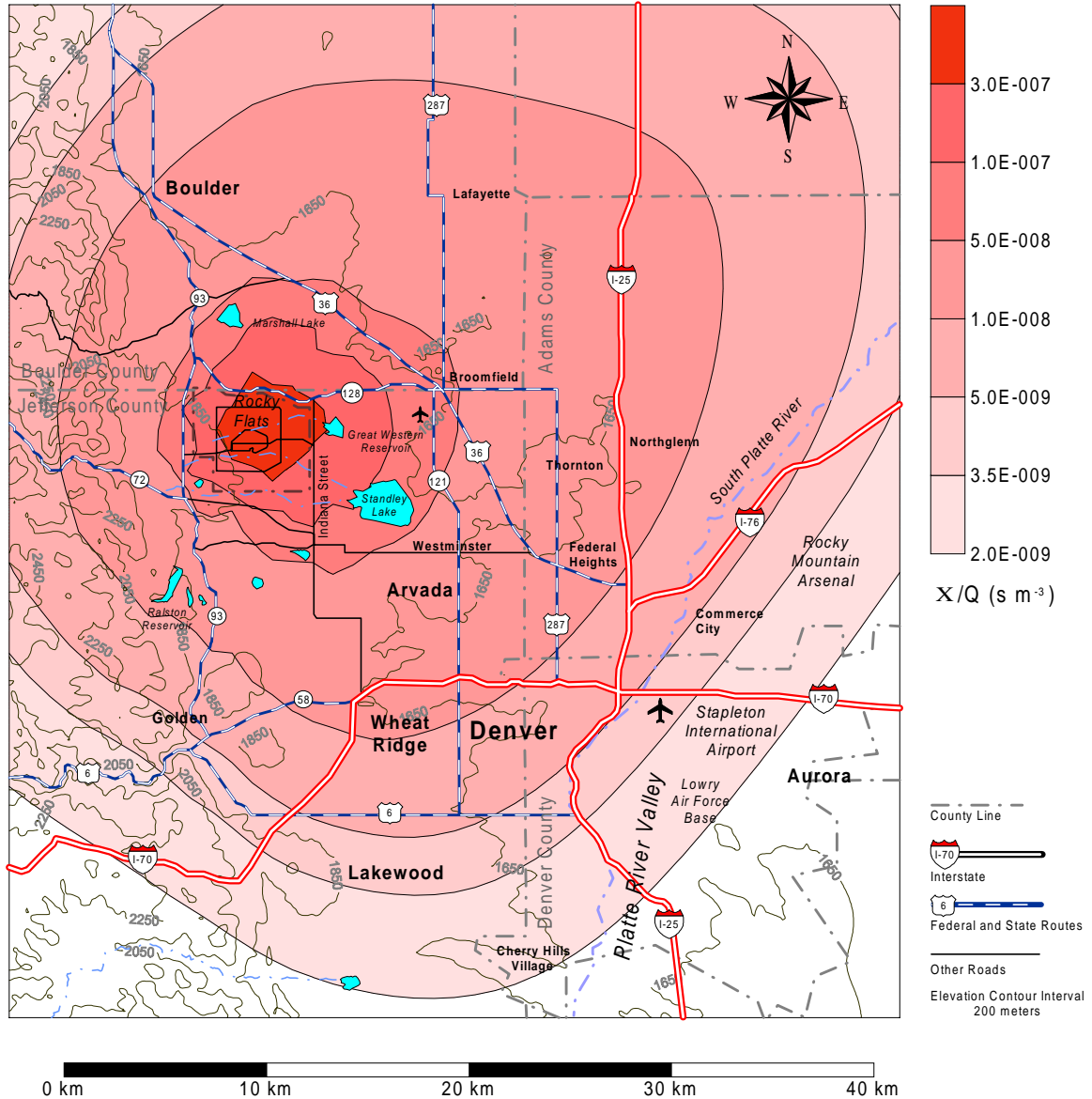
Deposition dispersion factors were also calculated for continuous release sources. The deposition dispersion factor is the ratio of the total deposition (in curies per square meter) to the source release rate (in curies per year). These factors were combined with source release rates to derive ground-level deposition in the model domain from continuous sources. Deposition dispersion factors were calculated using RATCHET in a similar manner to the dispersion factors for concentration. The model includes contributions from both 903 Area baseline and routine sources. However, deposition from routine operations was minimal and often resulted in surface soil concentrations that were not distinguishable from background plutonium concentrations. Dispersion factors for deposition from 903 Area baseline sources (Figure 8) show a different pattern than air concentration dispersion factors because the deposition velocity increases with surface roughness ( $z_o$ ). The area of enhanced deposition in the foothills west of the site is in response to the higher roughness heights (2.0 m) assigned to the foothills compared to roughness heights assigned to remainder of the model domain (0.05–0.6 m).

### Time-Integrated Concentration and Deposition from Discrete Events

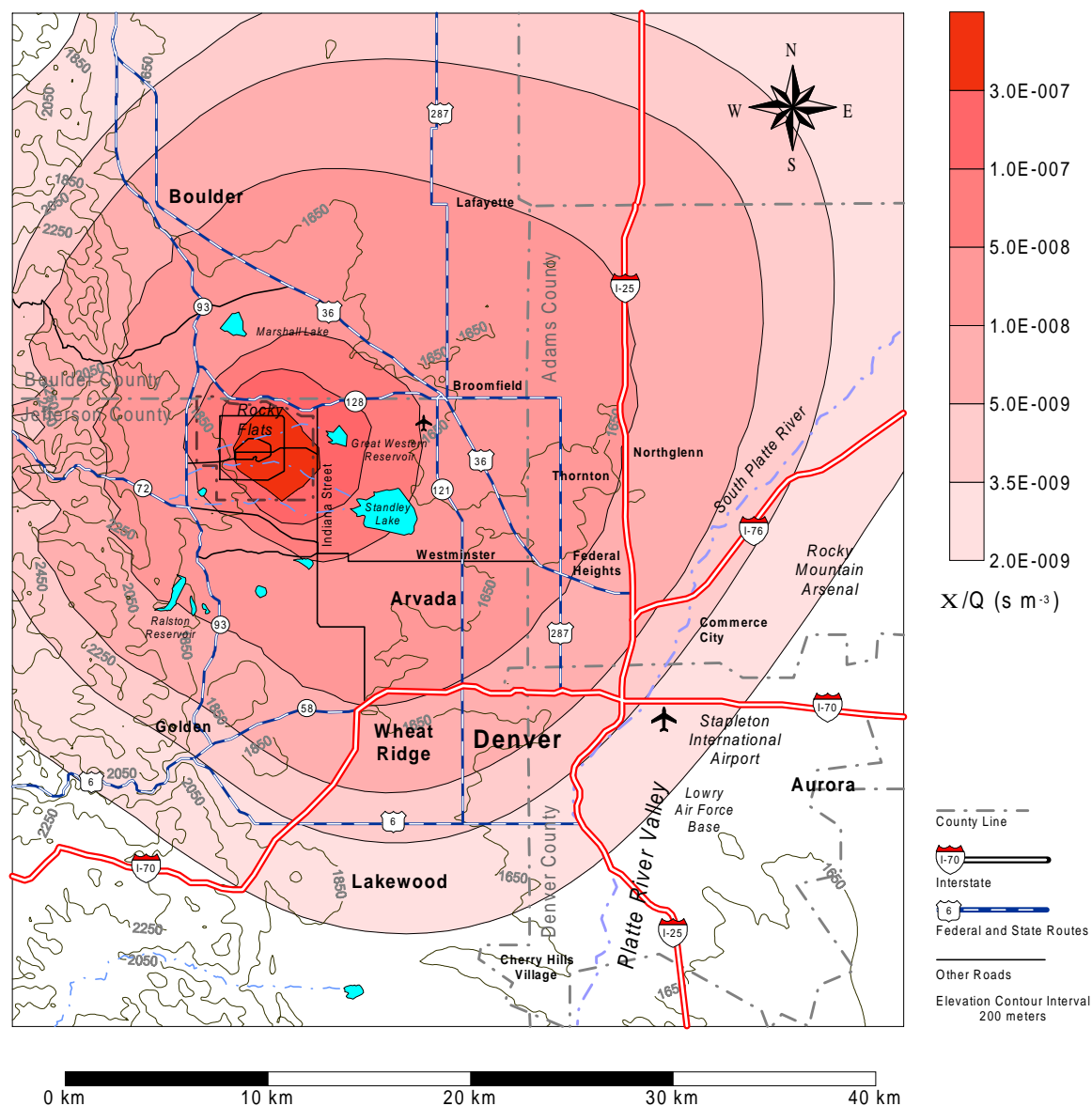
Time-integrated concentrations and deposition from discrete events are documented in Rood and Grogan (1999a), (1999b), and (1999c) for the 903 Area, 1957 fire, and 1969 fire releases, respectively. Concentrations were calculated using the RATCHET code and meteorological data for the specific days the events occurred. RATCHET was run using its Monte Carlo sampling features that sampled from distributions of basic transport parameters for each trial within a simulation. The source term was also sampled within the Monte Carlo simulation. Output consisted of  $n$  realizations of  $TIC$  and deposition at each of the 2295 computational nodes in the model domain. The computer code reads sequentially, concentrations and deposition at each node for each model realization. The number of hours that each discrete event occurred was also input (Table 3). The RATCHET model was run for several hours after releases ceased to allow the plume to fully dissipate from the model domain.



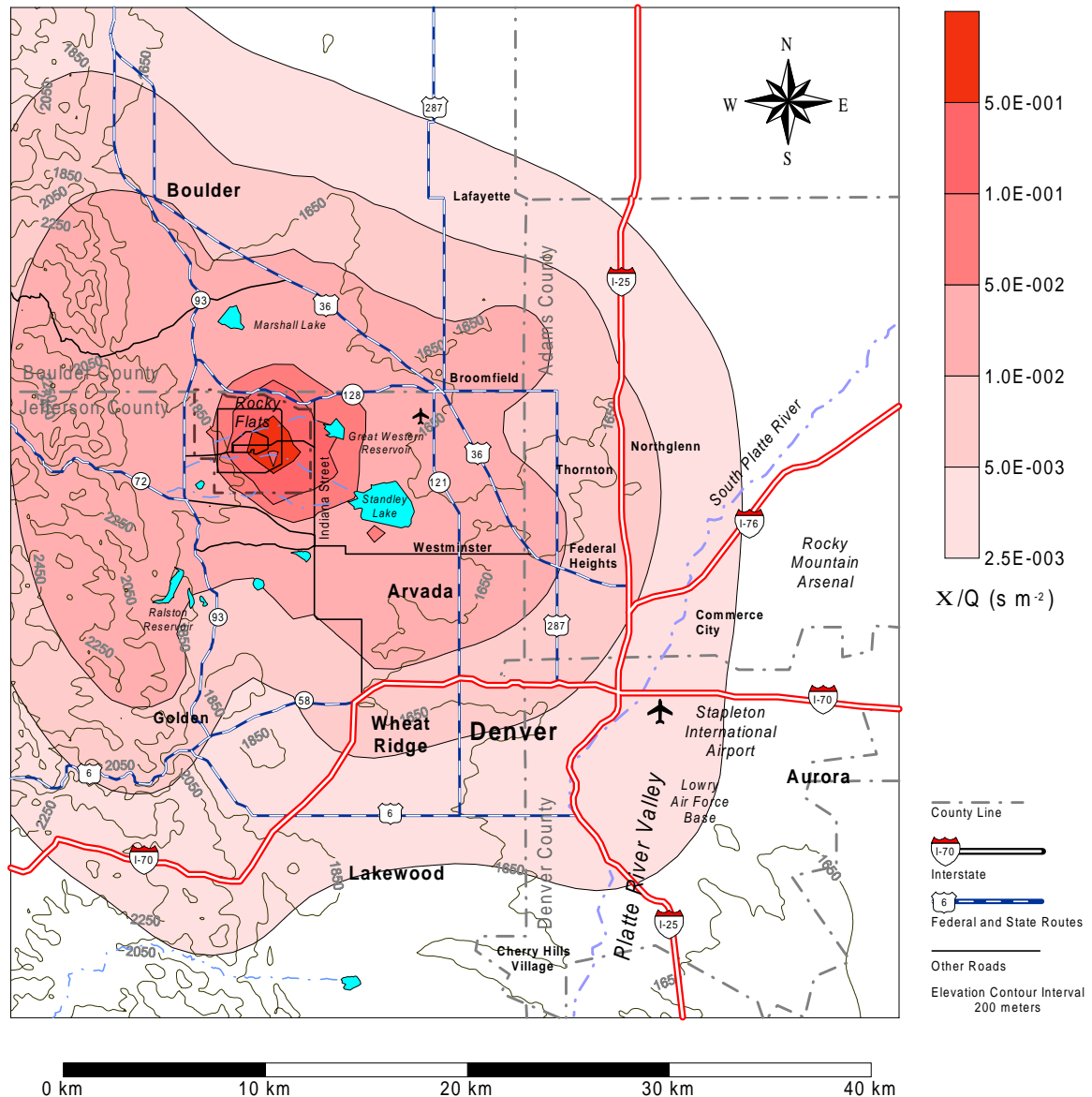
**Figure 5.** Isopleth map of annual average dispersion factors ( $X/Q$ ) for releases from the Building 771 stack using meteorological data from the Rocky Flats Plant and Denver Stapleton International Airport from 1989–1993.



**Figure 6.** Isopleth map of annual average dispersion factors ( $X/Q$ ) for releases from the Building 776 roof vents using meteorological data from the Rocky Flats Plant and Denver Stapleton International Airport from 1989–1993.



**Figure 7.** Isopleth map of annual average dispersion factors ( $X/Q$ ) for particulate releases  $<3 \mu\text{m}$  from the 903 Area using meteorological data from the Rocky Flats Plant and Denver Stapleton International Airport from 1989–1993.



**Figure 8.** Isopleth map of annual average deposition dispersion factors for baseline 903 Area releases using meteorological data from the Rocky Flats Plant and Denver Stapleton International Airport from 1989–1993. A mean particle size of  $20\ \mu m$  AED was used in the calculations.

**Table 3. Number of Hours Represented by Each Discrete Event**

Discrete event	Number of hours in model simulation
1957 Fire (September 11 and 12, 1957)	9
903 Area releases <sup>a</sup>	26
1969 Fire (May 11 and 12, 1969)	15

<sup>a</sup> Discrete 903 Area releases include December 5, 1968; January 6, 7, 30, 1969; March 19, 1969; and April 7, 1969.

Ground-level *TICs* at the 50th percentile level are illustrated in Figures 9 through 11 for the 1957 fire, 903 Area, and 1969 fire, respectively. The 903 Area plot includes contributions from the six discrete events that occurred in 1968 and 1969 and represents activity attached to respirable particles ( $<15\ \mu\text{m}$  AED). Tables 4 through 6 present the 5th, 50th, and 95th percentiles for ground-level *TICs* at selected receptor locations in the model domain for discrete events. The *TIC* values for the 1957 fire were greatest, followed by the 903 Area and 1969 fire. Note that plumes for the 1957 fire and 903 Area discrete releases trend easterly, while plumes for the 1969 fire trend east and west. These trends reflect the meteorological conditions that prevailed during each release. The largest releases from the 1957 fire occurred during the evening hours (10:00–11:00 p.m.). Downslope conditions, where air from the foothills moves easterly until it converges with an air mass moving down (northeast) the Platte River Valley, typically persist during this time (Crow 1974). During the evening of September 11th, 1957, surface winds were light and out of the north-northwest. Therefore, the contaminant plume had a south-southeast initial trajectory and then changed to the northeast as the air masses converged near the Platte River Valley. The easterly trajectories of 903 Area releases reflect the passage of synoptic weather fronts characterized by strong westerly winds. Releases from the 1969 fire occurred during the afternoon when upslope conditions persisted. Upslope conditions are a result of daytime heating and typically result in easterly winds that prevail during the daylight hours. The geometry of the plume mass reflects the transition from upslope to downslope conditions during the model simulation period.

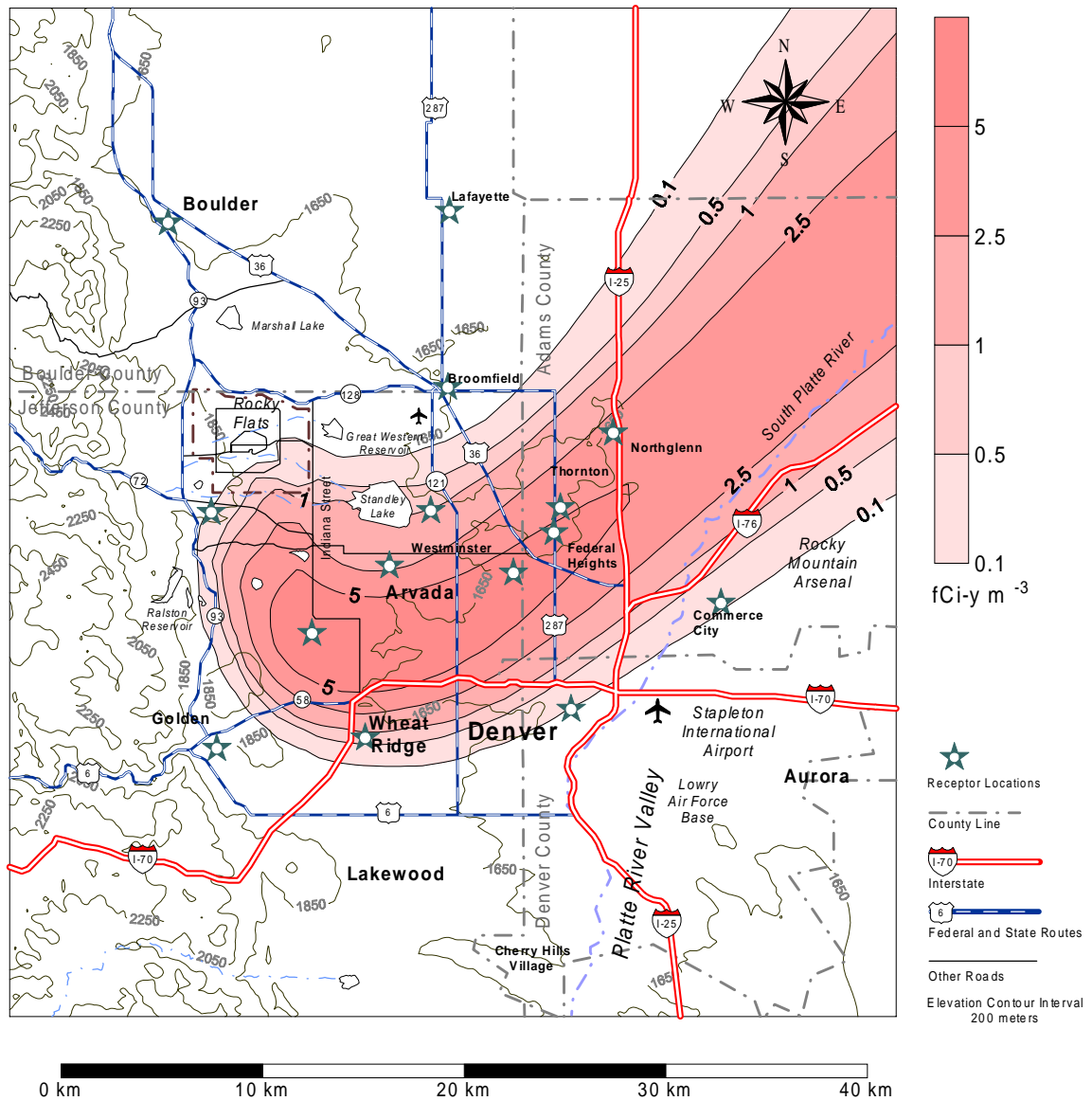
Surface deposition values (in curies per square meter) were input into the model in a similar manner to *TIC*. The 903 Area releases were by far the largest contributor to offsite plutonium contamination in soil because the larger particle sizes associated with the suspended activity had higher gravitational settling velocities. This greater settling velocity causes contamination to deposit relatively close to the source. Predicted soil concentrations in the 0–3-cm soil layer resulting from the 903 Area discrete events are illustrated in Figure 12. Surface deposition values (in curies per square meter) were converted to soil concentrations in the 0–3-cm layer (in picocuries per gram) using the depth distribution equations discussed later in this report and average soil density provided by Webb (1996).

## Resuspension Factors

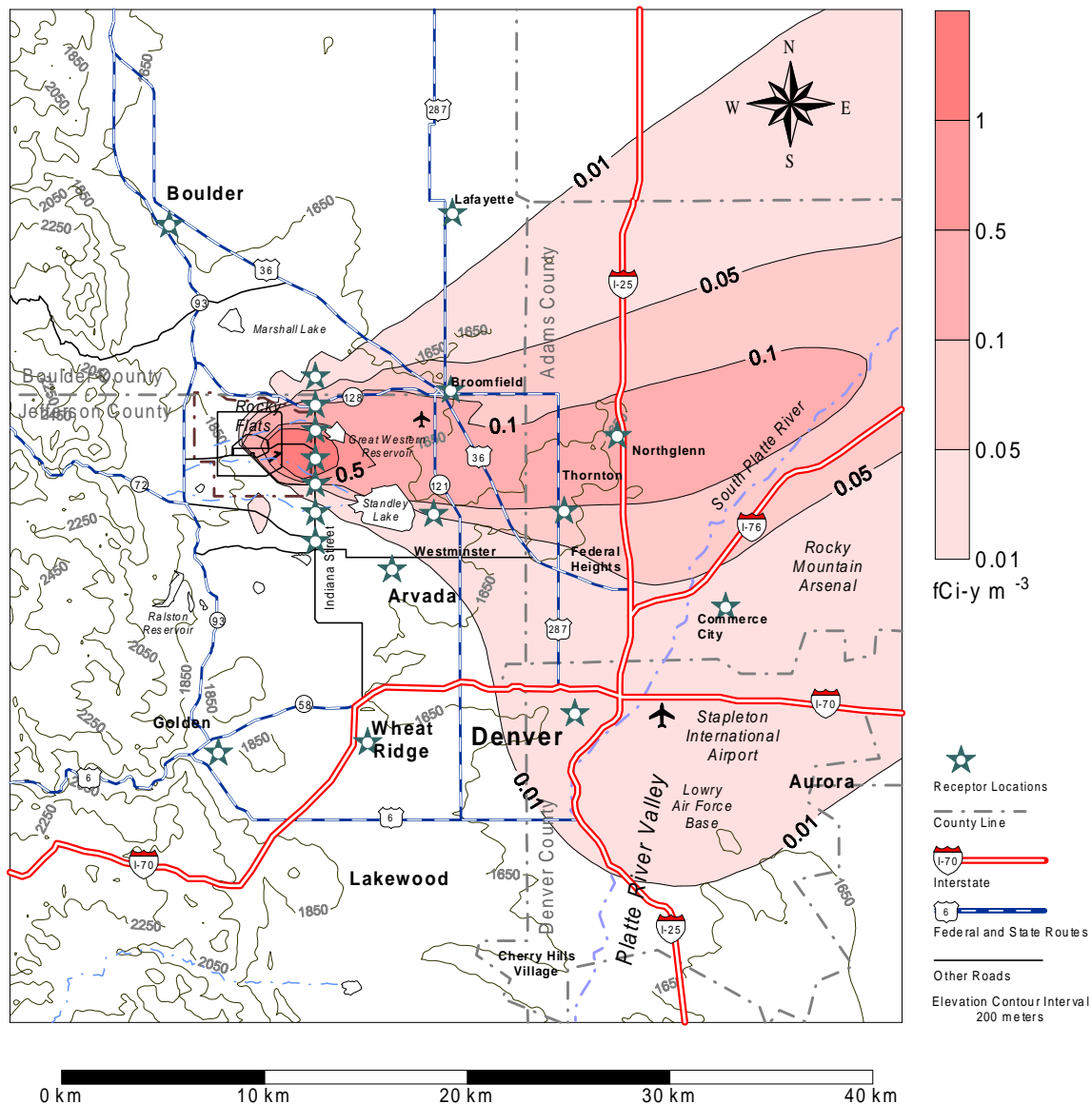
The model for resuspension (Equation 12) requires two resuspension factors. The first ( $A$  in Equation 12) represents the resuspension from newly deposited activity, and it decreases exponentially over time with a half-life of about 1 year. The second resuspension factor ( $B$  in Equation 12) represents the long-term behavior of contaminated soil, after weathering and redistribution of activity within the soil. The second factor is about 3 orders of magnitude smaller than the first. Suggested values for  $A$  and  $B$  were  $1 \times 10^{-6}\ \text{m}^{-1}$  and  $1 \times 10^{-9}\ \text{m}^{-1}$ , respectively.

Measured resuspension factors at Rocky Flats are summarized in Table 7. Note that the resuspension factor is sensitive to the sampling depth as illustrated by the data from Volchok. Little detail was provided about soil concentration measurements used to derive resuspension factor estimates in Sehmel and Orgill (1972) and Langer (1991). Sampling practices at the time used sampling depths ranging from 3–20 cm (Rope et al. 1999). For purposes of this calculation, we assumed resuspension factor measurements were based on the 0–3-cm layer.



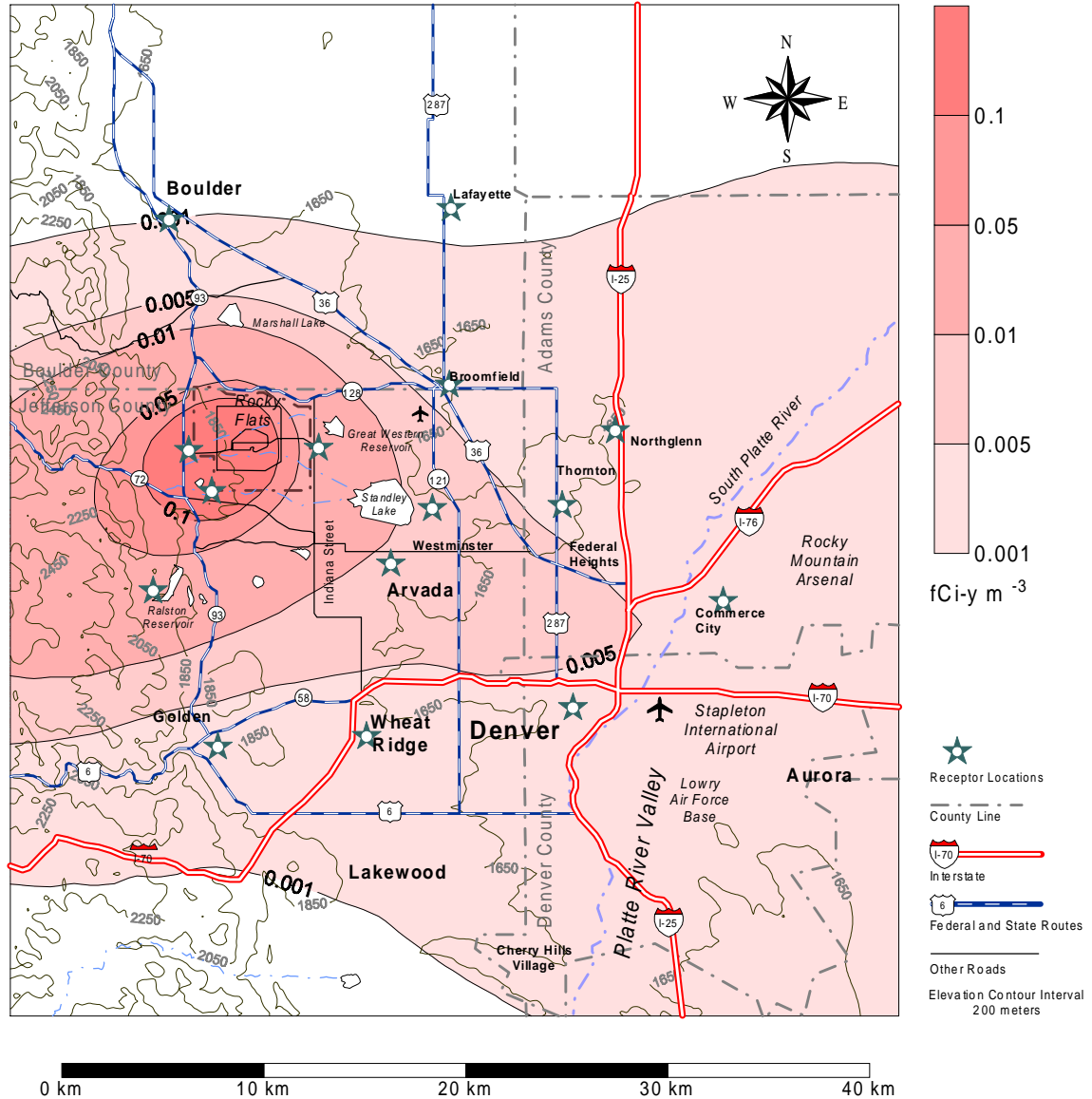


**Figure 9.** Ground-level time-integrated concentration of plutonium at the 50th percentile level for the 1957 fire. Receptor locations correspond to those listed in [Table 4](#).



**Figure 10.** Ground-level time-integrated concentration of plutonium activity attached to respirable particles (<15 μm AED) at the 50th percentile level for 903 Area discrete events. Receptor locations correspond to those listed in [Table 5](#).





**Figure 11.** Ground-level time-integrated concentration of plutonium at the 50th percentile level for releases from the 1969 fire. Receptor locations correspond to those listed in [Table 6](#).

**Table 4. Ground-Level Time-Integrated Concentrations at Selected Receptor Locations for the 1957 Fire (fCi-y m<sup>-3</sup>)**

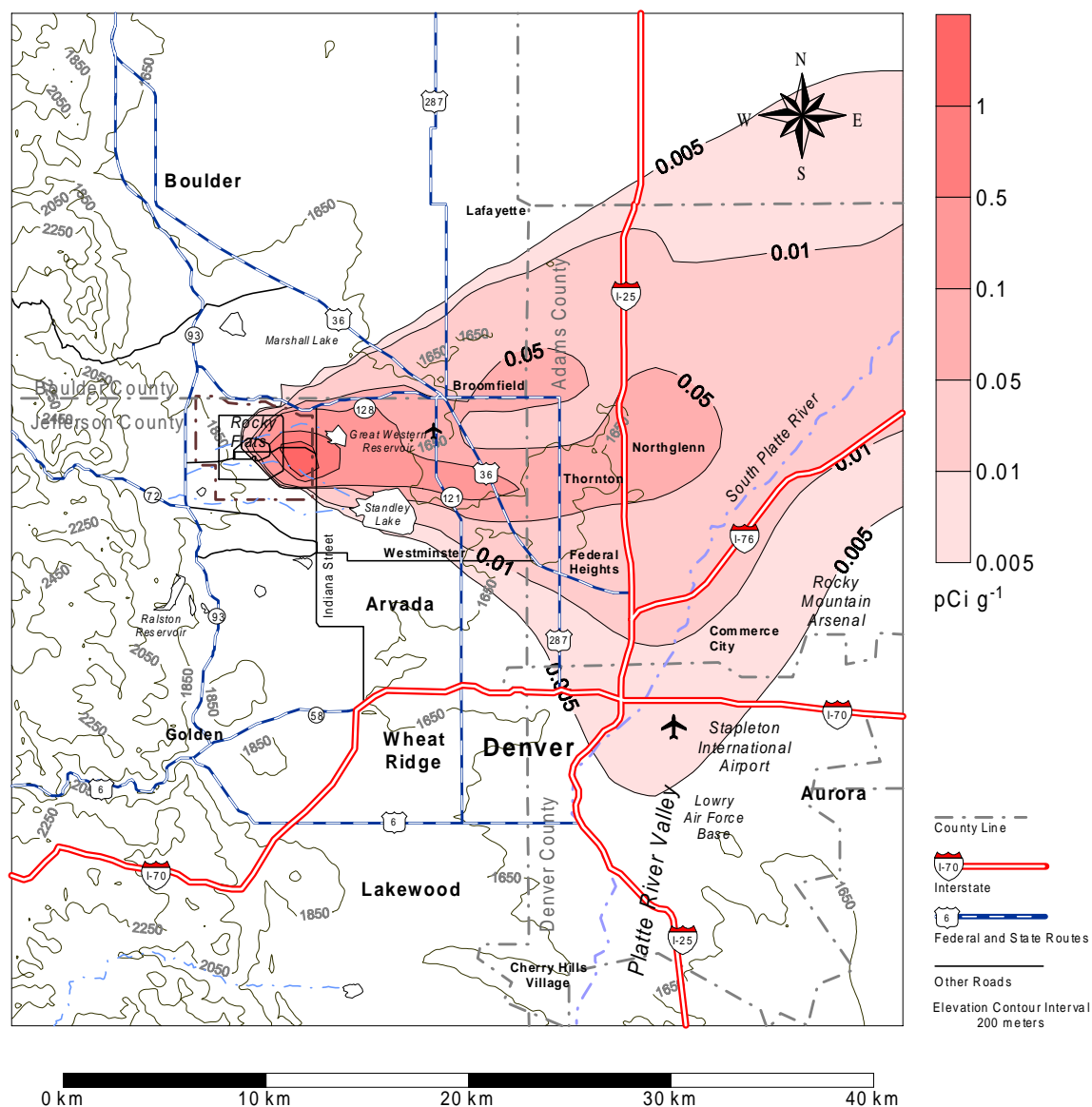
Receptor	UTM <sup>a</sup> E (m)	UTM N (m)	Percentile level		
			95th	50th	5th
Arvada East	495850	4409050	156.30	6.41	2.82
Federal Heights	497850	4411050	110.10	5.58	2.51
Arvada West	485850	4406050	451.20	8.14	0.09
So RFP Buffer	480850	4412050	1632.00	0.11	0.00
Arvada	489696	4409400	246.50	2.44	0.53
Wheat Ridge	488491	4400870	461.60	0.73	0.00
Westminster	491747	4412140	41.40	1.08	0.28
Broomfield	492593	4418250	0.41	0.00	0.00
Thornton	500804	4415990	20.12	3.28	1.37
Northglenn	498184	4412290	73.64	4.68	2.16
Boulder	478709	4426410	0.00	0.00	0.00
Lafayette	492674	4426980	0.02	0.00	0.00
Golden	481126	4400350	87.92	0.00	0.00
Commerce City	506152	4407550	5.45	0.09	0.00
Denver	498712	4402290	12.33	0.04	0.00

<sup>a</sup> UTM = universal transverse mercator.**Table 5. Ground-Level Time-Integrated Concentrations of Respirable Particles (<15 µm AED) at Selected Receptor Locations for 903 Area Discrete Releases (fCi-y m<sup>-3</sup>)**

Receptor	UTM E (m)	UTM N (m)	Percentile level		
			95th	50th	5th
Indiana Street	485912	4418920	0.10	0.015	0.003
Indiana Street	485912	4417520	1.7	0.24	0.038
Indiana Street	485912	4416280	5.2	1.2	0.29
Indiana Street	485912	4414870	9.1	3.0	1.2
Indiana Street	485912	4413630	0.38	0.091	0.031
Indiana Street	485912	4412220	0.018	0.006	0.002
Indiana Street	485912	4410790	0.008	0.004	0.002
Arvada	489696	4409400	0.013	0.006	0.002
Wheat Ridge	488491	4400870	0.01	0.004	0.001
Westminster	491747	4412140	0.12	0.040	0.014
Broomfield	492593	4418250	0.51	0.10	0.035
Northglenn	500804	4415990	0.48	0.17	0.068
Thornton	498184	4412290	0.19	0.080	0.031
Boulder	478709	4426410	0.005	0.001	0.000
Lafayette	492674	4426980	0.014	0.005	0.002
Golden	481126	4400350	0.010	0.002	0.001
Commerce City	506152	4407550	0.10	0.037	0.016
Denver	498712	4402290	0.054	0.017	0.006

**Table 6. Ground-Level Time-Integrated Concentrations at Selected Receptor Locations  
for the 1969 Fire (fCi-y m<sup>-3</sup>)**

Receptor	UTM E (m)	UTM N (m)	Percentile		
			95 <sup>th</sup>	50 <sup>th</sup>	5 <sup>th</sup>
SW Buffer Zone	480850	4413050	0.937	0.234	0.032
East Entrance	485850	4415050	0.208	0.035	0.004
West Entrance	479850	4415050	1.178	0.302	0.054
Ralston Reservoir	477850	4408050	0.103	0.027	0.004
Arvada	489850	4409050	0.028	0.008	0.002
Wheat Ridge	488850	4401050	0.012	0.003	0.000
Westminster	491850	4412050	0.024	0.008	0.002
Broomfield	492850	4418050	0.016	0.004	0.000
Thornton	500850	4416050	0.010	0.003	0.000
Northglenn	497850	4412050	0.013	0.005	0.001
Boulder	478850	4426050	0.010	0.001	0.000
Lafayette	492850	4427050	0.005	0.001	0.000
Golden	480850	4400050	0.014	0.003	0.000
Commerce City	505850	4407050	0.012	0.005	0.001
Denver	498850	4402050	0.012	0.004	0.001



**Figure 12.** Predicted plutonium soil concentrations in the 0–3-cm layer from 903 Area discrete events. Background soil concentrations are around 0.05 pCi g<sup>-1</sup>.

**Table 7. Resuspension Factors Measured at the Rocky Flats Plant**

Study	Year measured	Sampling depth (cm)	Resuspension factor ( $\text{m}^{-1}$ )
<a href="#">Sehmel and Orgill</a> (1972)	1970–1971	3–20 <sup>a</sup>	$10^{-5}$ to $10^{-9}$
Volchock <i>in</i> <a href="#">Linsley</a> (1978)	1972?	20	$10^{-9}$
Volchock <i>in</i> <a href="#">Linsley</a> (1978)	1972?	0.2	$10^{-6}$
<a href="#">Krey et al.</a> (1974) <sup>b</sup>	1973	3–20 <sup>a</sup>	$3 \times 10^{-9}$
<a href="#">Rockwell</a> (1985)	~1978	0.3	$1-2 \times 10^{-9}$
<a href="#">Langer</a> (1991)	1989–1990	3–20 <sup>a</sup>	$10^{-13}$ to $10^{-10}$

<sup>a</sup> Estimated sampling depth based on sampling practices at the time.

<sup>b</sup> This value was apparently not measured by Krey, but it was reported as the current estimate of the resuspension factor at the time of his study.

The resuspension factor measurements were all performed after 1970 in the field east of the 903 Area. Therefore, these estimates were dominated by deposition from 903 Area sources in 1968 and 1969. The decrease in the resuspension factor over time can roughly be observed in the data presented in Table 7. The measured resuspension factors reported by Sehmel represent short-term (6 hours to 1 week) estimates. Values representing annual average conditions will likely be lower. The data from Langer provide an estimate of the long-term resuspension factor. His values were based on several years of field measurements and are considerably lower than those reported in the literature. Both Sehmel and Langer comment on the deficiencies of the resuspension factor, pointing out its inadequacies in addressing the physical processes affecting resuspension and that airborne concentrations at Rocky Flats are more related to upwind sources and not local surface contamination. We acknowledge these deficiencies in the resuspension factor approach but find it difficult to move beyond such an approach at this time without additional model research and site-specific field investigation.

Based on the information provided in [Table 7](#) and the above discussion, we selected distributions for parameters *A* and *B* in the resuspension factor equation ([Equation 12](#), [Table 8](#)). We took the measurements made by [Sehmel and Orgill](#) (1972) to represent the initial, short-term resuspension factor (*A*) and those of the other researchers (Krey, Langer, Linsley, and Rockwell) to represent the long-term resuspension factor (*B*). Lognormal distributions were assigned to the short- and long-term resuspension factors. The geometric mean was assumed to be at the log-transformed midpoint of the range of measured data. The minimum and maximum resuspension factor was taken to represent the 1st and 99th percentile values of the distribution. The geometric standard deviation was then given by

$$GSD = \exp \left[ \frac{\ln(99th/1st)}{2t(\alpha/2, d)} \right] \quad (16)$$

where

*GSD* = geometric standard deviation

*t* = t distribution for *d* degrees of freedom (infinite) and probability of  $\alpha/2$  (0.01).

**Table 8. Distribution of Short-term (A) and Long-term (B) Resuspension Factors**

Parameter	Geometric mean	
	( $\text{m}^{-1}$ )	Geometric standard deviation
Short-term resuspension factor (A)	$3.2 \times 10^{-7}$	3.8
Long-term resuspension factor (B)	$3.2 \times 10^{-11}$	3.8

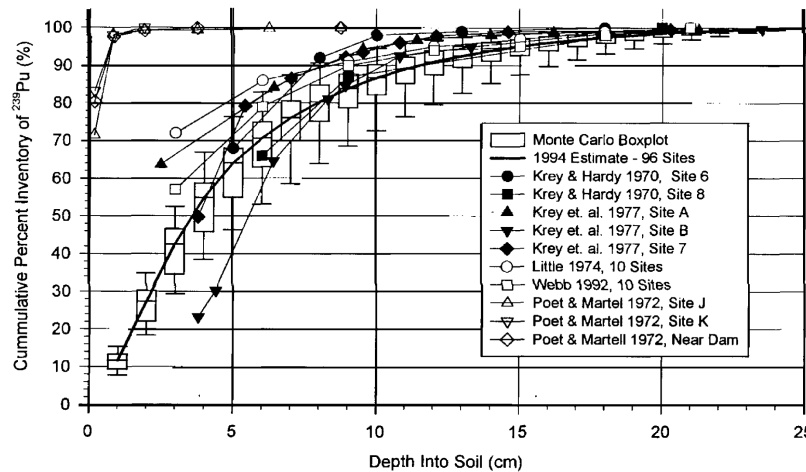
For the short-term resuspension factor, a minimum and maximum (1st and 99th percentile) was taken to be  $1 \times 10^{-8}$  to  $1 \times 10^{-5} \text{ m}^{-1}$ , respectively. For the long-term resuspension factor, a minimum and maximum was taken to be  $1 \times 10^{-12}$  to  $1 \times 10^{-9} \text{ m}^{-1}$ , respectively. We did not include the lower bound long-term resuspension factor measured by [Langer](#) (1991) because the Rockwell International data ([Rockwell](#) 1985) seemed to suggest long-term resuspension was somewhat higher.

The resuspension factor was applied to the 0–3-cm layer of soil. It is recognized that resuspension occurs from a much thinner layer, typically several millimeters. However, this makes little difference if the depth of sampling used to determine the resuspension factor is the same as the modeled depth used when applying the resuspension factor. The time-dependent formulation of the resuspension factor accounts for weathering of surface activity in the 0–3-cm layer and movement of activity within that layer. Plutonium depth profiles have indicated activity has migrated down to 20 cm. Movement of activity out of the 0–3-cm layer was modeled separately and is discussed in the next section.

### Plutonium Depth Distribution and Soil Density

Comparison of predicted-to-observed plutonium concentrations in soil requires that we average the plutonium across a sampling depth and divide by the soil density (see [Equation 8](#)). Unfortunately, sampling depths used in the soil sampling studies performed in the past ([Krey and Hardy](#) 1970; [Krey et al.](#) 1977; [Little](#) 1976; [Poet and Martell](#) 1972; Webb [1992](#), [1996](#)) were not consistent. Therefore, sampling depth is important to consider when comparing predicted to measured soil concentrations.

Field studies of plutonium concentrations in soil provide evidence that plutonium has migrated from the surface down to a depth of about 20 cm. [Webb](#) (1996) provides an excellent summary of these data and calculated the fraction of the total plutonium inventory in soil as a function of depth for each of the major studies ([Figure 13](#)). The first soil studies conducted in February 1970 by [Krey and Hardy](#) (1970) indicated plutonium had migrated beyond 13 cm below the surface. In another study conducted about the same time, [Poet and Martell](#) (1972) measured plutonium depth distributions at seven sites east of the 903 Area and concluded that most of the plutonium remained in a 0–1-cm layer, which contradicted Krey and Hardy's findings. They argued that the larger fraction of plutonium found at depth in [Krey and Hardy](#) 1970 study was at sites distant from the heavily contaminated 903 Area and in areas disturbed by human activity. Sites measured by [Krey and Hardy](#) (1970) and reported in [Webb](#) (1996) were located east of the 903 Area and within the original buffer zone. Most of the contamination at these locations is thought to be of 903 Area origin. It is unknown whether these sites were disturbed. [Krey and Hardy](#) also measured depth profiles at six other sites; however, three of the sites were at distances greater than 32 km from the 903 Area. The distant sites showed a significant drop in the percent inventory in the 0–5-cm-layer (39–45%). At one site near Great Western Reservoir



**Figure 13.** The distribution of  $^{239}\text{Pu}$  with depth in soil near Rocky Flats as reported in several previous studies and by Webb (reproduced from [Webb 1996](#)).

(Site 12), 91% of the inventory was in the 0–5-cm layer. When the three distant sites were omitted, the mean of the percent inventory in the 0–5-cm depth was 72%, with a standard error of 6.7. This mean value excluded one site where the 0–5-cm depth was not measured. The mean value of the four sites is more in line with what [Little \(1976\)](#) measured in 1974. Using 10 sites, Little measured about 72% of the plutonium inventory in the 0–3-cm layer.

Krey's later work ([Krey et al. 1977](#)) was performed between 1973 and 1976. Depth profiles at three sampling sites were analyzed, and an empirical model was developed that described the movement of plutonium down the soil column with time. The model assumes loss by first-order processes down to a depth ranging from 3–7 cm. Beyond that depth, diffusion is the dominant transport process. The three sites were all within the plant buffer zone. Two of the sites (A and B) were located about 75 m south of the 903 Area and the third site (7) was located about 250 m southeast of the 903 Area near Woman Creek. Sites A and B showed substantially different profiles despite their close proximity to one another. However, Site B was reported to be located on a steep slope, while Site A was located on more level ground. This may account for the lower inventory in the surface because erosion may have removed activity from the surface at Site B.

In another study, [Jones and Zhang \(1994\)](#) used data collected by the Colorado Department of Health Radiation Control Division from 1970 to 1991 to examine the spatial and temporal trends of plutonium in soil around Rocky Flats. They found no evidence that surface soil concentrations were decreasing with time. Sampling depths for this study, however, were shallow and increased over time. In 1970, the sample depth was 0.16 cm and in 1989, the sample depth was 0.64 cm. [Jones and Zhang](#) attributed the observed decrease in soil concentration over time to dilution in samples that were collected at greater depth as the survey technique evolved over time.

Results from these studies are perplexing. There appears to be a clear evidence of a decrease in the 0–3-cm plutonium inventory between 1972 and 1989 based on the work of [Little \(1976\)](#), [Webb \(1992\)](#), and one sampling site in [Krey et al. \(1977\)](#). However, two of the other sites measured by [Krey et al. \(1977\)](#) show substantially less plutonium in the surface (0–5 cm) than was observed by Webb and Little. [Little \(1976\)](#) measured depth profiles at 10 sites and [Webb](#)

(1992) resampled these same sites in 1989, while Krey's later measurements were from only three sites.

It is unreasonable to assume instantaneous migration of plutonium down the soil column after deposition. Numerous processes discussed earlier can influence plutonium migration in the subsurface, and these processes are both temporally and spatially variable. Recent work by I. Litaor has suggested that under saturated soil conditions, plutonium can migrate very rapidly. This work is currently unpublished; however, it suggests that certain discrete events (such as heavy rainfall) may have moved plutonium into the subsurface in a relatively short period of time. For the remainder of time, plutonium has migrated very little.

Based on the summary of depth profiles provided in [Webb \(1996\)](#), we have fitted an equation that describes the temporal variation of the plutonium in the 0–3-cm soil layer ([Figure 14](#)). In doing so, we made the following assumptions and applied the following criteria:

- Plutonium soil data used in the regression reflect mainly deposition from 903 Area releases
- Most of the plutonium deposition occurred in early 1969
- One-hundred percent of the soil inventory was in the 0–3-cm layer in early 1969
- Sampling studies that did not measure the 0–3-cm soil layer were omitted from the analysis.

Using the assumptions and criteria stated above, we performed regression on six data points that yielded the following equation ( $r^2 = 0.972$ )

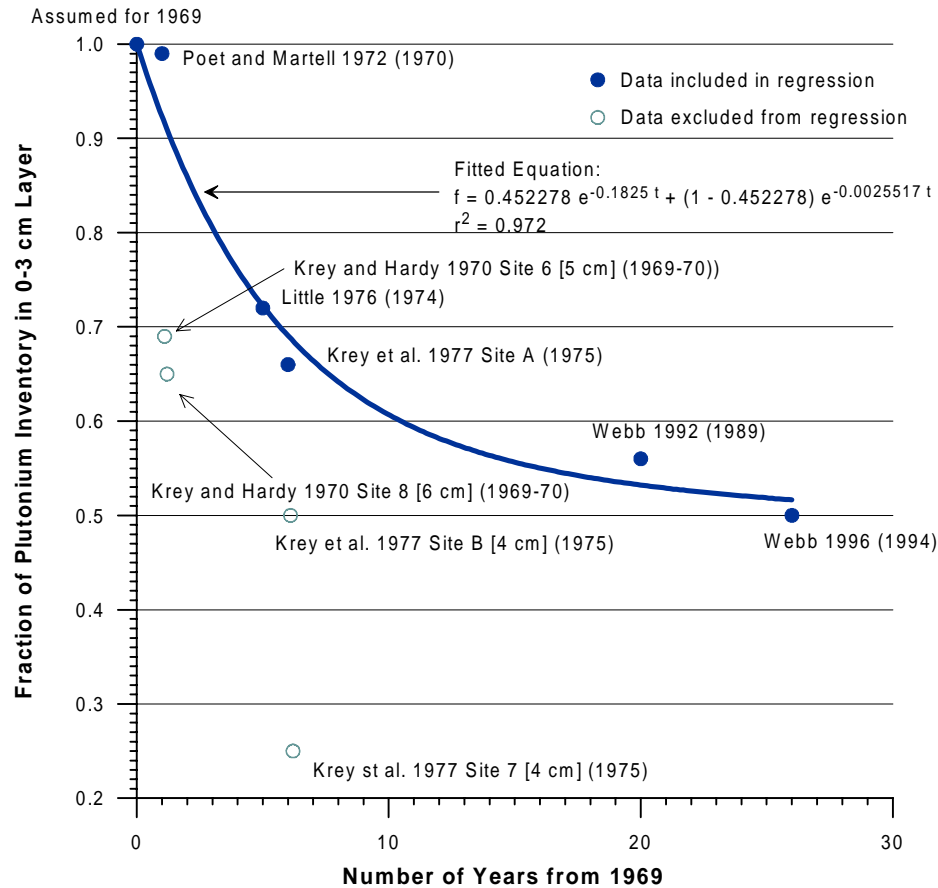
$$f(t) = 0.452278 e^{-0.1825t} + (1 - 0.452278) e^{-0.0025517t} \quad (17)$$

Equation (17) provides the fraction of plutonium in the 0–3-cm layer as a function of time following a surface deposit at  $t = 0$ . The fraction of plutonium inventory in the 0–3-cm layer from transient deposition is found using the convolution integral ([Equation 14](#)).

Equation (17) omitted much of Krey's data. Sites 6 and 8 in [Krey and Hardy \(1970\)](#) were not included in the regression because minimum sampling depths were  $>3$  cm. In later work ([Krey et al. 1977](#)), Krey measurements indicated between 25 to 75% of the plutonium inventory was in the first 4 cm of soil based on three sampling sites. However, only one of the three sites sampled down to 3 cm.

One may argue that we have omitted critical data from the analysis because it would have made for poor regression. We acknowledge this deficiency in our assessment, but we find it difficult to reconcile the few measurements made by Krey with over 116 measurements from Little and Webb that show a clear decrease in surface soil plutonium inventory over time. The net effect of omitting Krey's data points in the regression is that surface soil concentrations and subsequent resuspension might be overestimated during the 1970s. Given the fact that most of the plutonium exposure was from site releases that occurred before 1970, the relative importance of exposure from resuspension is probably small. There is no question that the depth distribution of plutonium in soil will vary spatially in the model domain. Equation (17) provides a quantitative description of the gross behavior of plutonium in soil. We make no attempt to quantify the behavior on a mechanistic level.





**Figure 14.** Regression of the percent of total plutonium inventory in the 0–3-cm soil layer as a function of time. The year in parenthesis was the year sampling took place. Data were obtained from the summary provided in [Webb \(1996\)](#).

[Webb \(1996\)](#) provides a detailed investigation of soil density around Rocky Flats. Webb studied the relationship between soil density with depth and the effects of the rock volume on estimated density. He argued that the rock volume should be excluded from soil density estimates because the rocks (derived from foothills erosion) do not contain plutonium in their mineral matrix. We agree with his assessment and have used his estimates of soil density in this work. Soil density was measured at each of Webb’s 96 sampling sites. These data were regressed yielding the following equation for soil density ( $\rho$ ) as a function of depth.

$$\rho = 0.79 d^{0.24} \quad (18)$$

The average soil density in the sampling layer was determined by integrating Equation (18) across the sampling depth and then dividing by that depth. The average density in the 0–3-cm surface layer was calculated to be  $0.829 \text{ g cm}^{-3}$ .

## Risk Coefficients

Estimates of risk coefficients are required to calculate the lifetime cancer incidence risk. Risk coefficients relate the lifetime risk of cancer incidence to the amount of plutonium inhaled.

Plutonium risk coefficients were developed in this phase of the study and are documented in [Grogan et al. \(1999\)](#).

The principal plutonium isotopes of concern at Rocky Flats are  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , which have long half-lives of 24,065 years and 6537 years, respectively. Plutonium emits alpha particles that are relatively heavy and slow, thus, creating short, dense ionization trails. Alpha particles have such weak penetration abilities that they can be blocked by a piece of paper, or the dead, outer layers of the skin. As a result, the major danger from plutonium comes from having it inside your body. For residents in the vicinity of Rocky Flats, plutonium is most likely to have entered the body from breathing air that contained plutonium particles released from the site. After inhalation, plutonium enters the blood and about 80% is transported to the bone or liver where it is retained for years. Following inhalation, the four most highly exposed tissues are bone surface, lung, liver, and bone marrow. These account for more than 97% of the total dose received by infants and adults alike. The dose per unit activity inhaled varies for these four tissues ([Table 9](#)). Furthermore, the dose per unit activity (dose conversion factor) also varies depending on the particle size distribution of the inhaled plutonium aerosol ([Table 9](#)). Three different particle size distributions are used to characterize the plutonium releases from the 903 Area at Rocky Flats (1, 5, and 10- $\mu\text{m}$  activity median aerodynamic diameter [AMAD] particles). Each of these distributions is assumed to be lognormal with a GSD of 2.5; therefore, each size distribution covers a relatively large range of particle sizes. The 1- $\mu\text{m}$  AMAD particle size distribution results in the largest doses to the tissues per unit intake of activity. This is because the particles penetrate deeper into the lungs and are retained longer. In all cases, the plutonium is assumed to be in the oxide form.

The incidence of health effects depends on the amount of dose received. There are two main classes of health effects induced by ionizing radiation: deterministic and stochastic effects. Deterministic effects most often follow acute, high dose exposure. The severity of the effect increases with dose above the threshold dose. Below the threshold dose, the effect is not evident; however, subtle minor effects may occur. Deterministic effects cause direct damage to tissues and include effects that most often occur within days to weeks after exposure. For example, these effects can cause reddening of the skin, cataracts, hair loss, sterility, and bone marrow depression after external irradiation. After inhalation of plutonium, deterministic effects may include radiation pneumonitis, pulmonary fibrosis, and lymphopenia, but these conditions occur only after very high doses. The threshold dose for most deterministic effects is at least 0.5 Gy delivered in a short time, and many threshold doses are much higher ([NCRP 1991](#)). For the releases of plutonium that occurred from the site, doses to individuals in the Rocky Flats area were well below the threshold doses. Therefore, deterministic health effects were not possible.

Stochastic effects are assumed to occur randomly at all dose levels, including the lowest doses. The frequency of stochastic effects is dependent on the dose, and the effects usually occur at long intervals after exposure. In a large population exposed to low doses, only a few of the exposed individuals will be affected, most will not. The two principal types of stochastic effects are induced cancer and genetic effects. For exposure to plutonium, the risk of induced cancer is the health effect of most concern; in particular, lung cancer, liver cancer, bone cancer, and leukemia (bone marrow exposure) are the induced cancers of concern because these tissues receive the highest doses. People exposed to radiation are several times more likely to be affected by an induced cancer than to transmit genetic effects to their children, and the plutonium doses to the gonads (ovaries or testes) are small compared to other organs of the body (40 times less than

the lung). Therefore, genetic effects are not an important risk for plutonium exposures and we did not consider them further.

**Table 9. Summary of Plutonium Oxide Inhalation Dose Conversion Factors<sup>a</sup>**

Cancer site	Dose conversion factor ( $\mu\text{Gy Bq}^{-1}$ ) <sup>b</sup>		
	1- $\mu\text{m}$ AMAD particles GSD = 2.5	5- $\mu\text{m}$ AMAD particles GSD = 2.5	10- $\mu\text{m}$ AMAD particles GSD = 2.5
Lung	4.4 (1.9)	2.6 (2.7)	1.2 (4.3)
Liver	2.0 (3.0)	0.95 (3.5)	0.42 (4.5)
Bone	9.0 (3.0)	4.6 (3.5)	2.1 (4.5)
Bone marrow	0.46 (3.0)	0.22 (3.5)	0.11 (4.5)

<sup>a</sup>. Values for 1- $\mu\text{m}$  AMAD from [ICRP 1995](#); 5 and 10- $\mu\text{m}$  were calculated in [Grogan et al. 1999](#)

<sup>b</sup> Geometric mean (geometric standard deviation).

The alpha particles emitted from plutonium are densely ionizing, and the linear energy transfer (LET) to the tissue is high over the short range (about 40  $\mu\text{m}$ ) of the alpha particles (thus, the name high-LET radiation). Other radiations, such as gamma rays and x-rays, are less densely ionizing and are termed low-LET radiations. The biological effects of low-LET radiation are better known than those of high-LET radiation. The differences between radiation types are important to the analysis because high-LET radiations are more biologically effective per unit of dose than low-LET radiations. This difference in effectiveness is usually described by the relative biological effectiveness (RBE), which is defined as the ratio of doses from two different radiations to produce the same type and level of biological effect.

Inhalation of plutonium results in the exposure of organs to high-LET radiation. While a few human populations have been exposed directly to large amounts of plutonium and some populations to other radionuclides that emit alpha particles, more groups have been exposed to low-LET gamma radiation and they have been evaluated in more epidemiologic detail. In addition, studies of cancer in animals exposed to both types of radiation and laboratory studies of cellular and other biological endpoints can be used to support human studies. We used these different sources of information in this phase of the study to develop four independent approaches to estimate the risk of cancer from radiation doses from plutonium deposited in the organs of the human body ([Grogan et al. 1999](#)). Three approaches used epidemiologic studies of human populations to derive dose-response relationships, and the fourth study used dose-response relationships from controlled animal experiments. We used the four independent approaches to derive, where possible, risk coefficients for each organ of interest. We then combined the coefficients from the different approaches by weighting each according its intrinsic merit to produce a single risk coefficient with uncertainties for each organ of interest.

We adjusted the overall mortality risk estimate for each cancer site by the lethality fraction to provide lifetime risk estimates for cancer incidence. The influence of gender and age was accounted for in the analyses (see [Grogan et al. \[1999\]](#) for details). The data allowed a distinction to be made between the risks and uncertainties to those under 20 years of age at exposure compared to those 20 and older. The data did not warrant a more detailed analysis. For this reason, the risk coefficients for persons under 20 years of age were applied to the infants and children in the seven hypothetical exposure scenarios.

The GM (50th percentile) and GSDs of the cancer incidence risk coefficient distributions are listed in Table 10. The units reported in [Grogan et al. \(1999\)](#) have been changed from risk per 100,000 persons per unit of activity in kilobecquerels (kBq) to risk per 10,000 persons per unit of activity in microcuries ( $\mu\text{Ci}$ ). These numbers indicate the median number of cases of cancer (fatal and nonfatal) that would be expected to result from 10,000 people all inhaling 1  $\mu\text{Ci}$  of  $^{239/240}\text{Pu}$  particles with the defined particle size distribution.

**Table 10. Lifetime Cancer Incidence Risk Per 10,000 Persons Per 1  $\mu\text{Ci}$  of Inhaled  $^{239/240}\text{Pu}$  for Three Particle Size Distributions**

1- $\mu\text{m}$ AMAD particles (GSD = 2.5)			
Cancer site	Gender	Under 20 <sup>a</sup>	20 and older <sup>a</sup>
Lung	Male	206 (3.5)	210 (3.4)
	Female	206 (3.5)	210 (3.4)
Liver	Male	92 (5.2)	49 (5.2)
	Female	45 (5.4)	23 (5.4)
Bone surface	Male	16 (9.5)	8.0 (9.3)
	Female	8.0 (10)	4.0 (10)
Bone marrow	Male	2.4 (6.1)	2.3 (6.3)
	Female	2.4 (6.1)	2.3 (6.3)
5- $\mu\text{m}$ AMAD particles (GSD = 2.5)			
Cancer site	Gender	Under 20	20 and older
Lung	Male	117 (4.3)	119 (4.2)
	Female	117 (4.3)	119 (4.2)
Liver	Male	46 (5.8)	24 (5.7)
	Female	21 (6.0)	11 (6.0)
Bone surface	Male	8.3 (10)	4.3 (10)
	Female	4.1 (11)	2.1 (11)
Bone marrow	Male	1.1 (6.7)	1.1 (6.7)
	Female	1.1 (6.7)	1.1 (6.7)
10- $\mu\text{m}$ AMAD particles (GSD = 2.5)			
Cancer site	Gender	Under 20	20 and older
Lung	Male	55 (6.1)	56 (6.0)
	Female	55 (6.1)	56 (6.0)
Liver	Male	21 (6.7)	11 (6.8)
	Female	9.6 (7.0)	5.0 (6.9)
Bone surface	Male	4.0 (12)	2.1 (12)
	Female	2.0 (12)	1.0 (12)
Bone marrow	Male	0.54(7.9)	0.54 (8.0)
	Female	0.54(7.9)	0.54 (8.0)

<sup>a</sup> Geometric mean (geometric standard deviation).

## Receptor Exposure Scenarios

One of the key parts of the Rocky Flats dose reconstruction work is calculating health impacts to people living in the surrounding area from materials released during RFP past operations. Dose reconstruction uses a pathways approach to study the potential radiation doses and health risks of these past releases on the surrounding communities. The pathways approach begins with learning what kinds of and how much material was released from a facility (source

terms) and ends with estimating the health impacts these releases had on the residents in the area. The risk to a person from exposure to the plutonium released depends upon a number of factors, such as

- Where the person lived and worked in relation to the RFP
- When and how long that person lived near the RFP (for example, during the key release events in 1957 and late 1960s or in the 1970s when releases were smaller)
- The age and gender of the person
- Lifestyle (that is, did the person spend a great deal of time outdoors or doing heavy work on a farm).

Although it is not realistic to calculate individual risks for every resident who may have lived or worked in the Rocky Flats area during its operational history, it is not credible to calculate a single risk that applies to all residents. To consider the many factors that influence exposure, we developed profiles, or exposure scenarios, of hypothetical but realistic residents of the RFP area for which representative risk estimates could be made. Each scenario represents one individual. These scenarios incorporate typical lifestyles, ages, genders, and lengths of time in the area. These scenarios can help individuals determine risk ranges for themselves by finding a lifestyle profile that most closely matches their background. The scenarios are not designed to include all conceivable lifestyles of residents who lived in this region during the time of RFP operations. Rather, they provide a range of potential profiles of people in the area and are designed to demonstrate the risk methodology.

The models and methodology presented earlier were demonstrated using five hypothetical exposure scenarios (Table 11). The individuals represented by these exposure scenarios were located at each receptor node model domain (see [Figure 3](#)), although some receptor locations are clearly not credible. Nevertheless, this methodology is useful in understanding the spatial dependency of exposure and risk. Each hypothetical receptor was assumed to spend all their exposure time at that location. The model accounts for some fraction of time spent out of the model domain. However, all receptors were assumed to be exposed to any discrete events that occurred during their exposure period.

As discussed earlier, inhalation was the only exposure pathway considered in this assessment. Ingestion of plutonium in water, food, and soil are potential pathways that could have been considered in more detail. However, plutonium compounds are very insoluble and tend to adhere to soil, making them relatively immobile and not readily taken up by plants or accumulated in the edible portions of animal products.

**Table 11. Exposure Scenario Descriptions**

Exposure scenario	Gender	Year of birth	Year beginning exposure	Year ending exposure	Days per year exposed
Laborer	Male	1934	1953	1989	365
Homemaker	Female	1934	1953	1989	350
Office worker	Female	1940	1965	1989	350
Infant-child	Female	1953	1953	1960	350
Student	Male	1957	1964	1974	350

The five exposure scenarios described in Table 11 were organized according to occupational and nonoccupational activities. Occupational activities included work, school, and extracurricular activities away from the home. Nonoccupational activities included time spent at home doing chores, sleeping, and leisure activities (such as watching television). In these calculations, the receptor was assumed to perform occupational and nonoccupational activities at the same location. The age of the receptor during which exposure occurred was also considered when calculating risk.

Each exposure scenario was divided into three types of activities: sleeping, nonoccupational, and occupational activities. For the infant-child scenario, occupational and nonoccupational activities are irrelevant; instead, activities were divided into sleeping and two other activities based on the child's age. For the infant, the other two activities were awake sedentary and awake active. For the child scenario, the two other activities were time spent at home (indoors and outdoors) and time at preschool or day care.

For each activity, we assigned time spent at four different exercise levels: resting, sitting (sedentary), light exercise, and heavy exercise. Some examples of light exercise are laboratory work, woodworking, housecleaning, and painting. Heavy exercise corresponds to occupations such as mining, construction, farming, and ranching. For each exercise level, we assigned an age- and gender-specific breathing rate. Breathing rates ([Table 12](#)) for persons age 8 and higher were obtained from [Roy and Courtay](#) (1991) and for children age 0–7 from [Layton](#) (1993).

Time budgets for various receptor activities were also based on [Roy and Courtay](#) (1991) ([Table 13](#)). The fraction of time spent at a specific exercise level while engaged in a given activity was assigned based on the nature of the activity. For example, the fraction of time spent at the resting exercise level while the receptor slept would be 1.0 and the other exercise levels would be 0. We applied a weighted-average breathing rate to each activity based on the number of hours spent at each exercise level. For the homemaker scenario, we separated nonoccupational activities into those performed indoors and outdoors. Although no distinction was made between indoor and outdoor air concentrations, exercise levels for indoor and outdoor activities differed. We calculated a time-weighted average breathing rate that included indoor and outdoor activities and applied it to nonoccupational time.

We calculated time-weighted average breathing rates for the three activities for which each receptor was assumed to be engaged. The time-weighted average breathing rate is given by

$$WBR_j = \sum_{i=1}^4 BR_i f_{i,j} \quad (19)$$

where

- $WBR_j$  = time-weighted average breathing rate for the  $j^{th}$  activity ( $\text{m}^3 \text{h}^{-1}$ )
- $BR_i$  = breathing rate for the  $i^{th}$  exercise level ( $\text{m}^3 \text{h}^{-1}$ )
- $f_{i,j}$  = fraction of time spent at the  $i^{th}$  exercise level for the  $j^{th}$  activity.

**Table 12. Breathing Rates for Various Exercise Levels as Reported in [Roy and Courtay](#) (1991) and [Layton](#) (1993)**

Gender	Age	Exercise level			
		Resting (m <sup>3</sup> h <sup>-1</sup> )	Sitting (m <sup>3</sup> h <sup>-1</sup> )	Light (m <sup>3</sup> h <sup>-1</sup> )	Heavy (m <sup>3</sup> h <sup>-1</sup> )
Male	30–60	0.45	0.54	1.50	3.00
Female	30–60	0.32	0.39	1.26	2.70
Male	18	0.50	0.60	1.58	3.06
Female	18	0.35	0.42	1.32	1.44
Male	16	0.43	0.52	1.52	3.02
Female	16	0.35	0.42	1.30	2.70
Male	15	0.42	0.48	1.38	2.92
Female	15	0.35	0.40	1.30	2.57
Male	14	0.41	0.49	1.40	2.71
Female	14	0.33	0.40	1.20	2.52
Male	12	0.38	0.47	1.23	2.42
Female	12	0.33	0.39	1.13	2.17
Male	10	0.31	0.38	1.12	2.22
Female	10	0.31	0.38	1.12	1.84
Male	8	0.29	0.39	1.02	1.68
Female	8	0.29	0.39	1.02	1.68
Male	3–7	0.24	0.29	0.72	1.68
Female	3–7	0.23	0.27	0.68	1.59
Male	0–3	0.19	0.23	0.58	1.35
Female	0–3	0.14	0.17	0.45	1.02

To summarize, we defined three activities for each exposure scenario: sleeping, occupational, and nonoccupational activities. The location of exposure for occupational and nonoccupational activities was assumed to be the same for all receptors. Four different exercise levels, each with an assigned breathing rate, were distinguished: resting, sitting, light exercise, and heavy exercise. The breathing rate during a given activity was the time-weighted average breathing rate of the four exercise levels. We also assumed breathing rates changed as a function of age.

The laborer scenario certainly represents the maximum exposed individual. The individual lived and worked in the model domain for the entire operational history of the site (1953–1989) and spent no time away from the model domain. The risks for this scenario were intended to represent upper-bound values. The other receptors were intended to represent more credible scenarios.



**Table 13. Time Budgets and Weighted Breathing Rates for the Exposure Scenarios**

Scenario	Activity	Fraction of time spent at a given activity level				Hours	Weighted breathing rate (m <sup>3</sup> h <sup>-1</sup> )
		Resting	Sitting	Light	Heavy		
Laborer	Occupational	0.00	0.00	0.25	0.75	8.0	2.625
	Nonoccupational	0.00	0.50	0.38	0.13	8.0	1.208
	Sleeping	1.00	0.00	0.00	0.00	8.0	0.450
	Weighted daily average						1.428
Office worker	Occupational	0.00	0.25	0.75	0.00	8.0	1.042
	Nonoccupational	0.00	0.50	0.38	0.13	8.0	1.004
	Sleeping	1.00	0.00	0.00	0.00	8.0	0.324
	Weighted daily average						0.790
Homemaker	Occupational	0.00	0.13	0.75	0.13	8.0	1.331
	Indoor nonoccupational	0.00	0.50	0.38	0.13	4.0	1.004
	Outdoor nonoccupational	0.00	0.38	0.50	0.13	4.0	1.113
	Total nonoccupational	0.00	0.44	0.44	0.13		1.058
	Sleeping	1.00	0.00	0.00	0.00	8.0	0.324
	Weighted daily average						0.904
Infant ( age 0-1 years) <sup>a</sup>	Awake—sedentary	0.00	0.71	0.29	0.00	7.0	0.254
	Awake—active	0.00	0.00	0.75	0.25	1.0	0.590
	Sleeping	1.00	0.00	0.00	0.00	16.0	0.144
	Weighted daily average						0.195
Child (age 2–6 years) <sup>a</sup>	Indoor (home)	0.00	0.50	0.42	0.08	6.0	0.549
	Outdoor (home)	0.00	0.00	0.67	0.33	1.5	0.980
	Total home					7.5	0.635
	Indoor (school)	0.00	0.80	0.20	0.00	2.5	0.351
	Sleeping	1.00	0.00	0.00	0.00	14.0	0.228
	Weighted daily average						0.368
Student (age 8) <sup>b</sup>	Indoor home	0.00	0.44	0.56	0.00	4.5	0.739
	Outdoor home	0.00	0.00	0.25	0.75	2.5	1.515
	Total home	0.00	0.22	0.40	0.38	7.0	1.016
	Indoor school	0.00	0.75	0.25	0.00	6.0	0.545
	Outdoor school	0.00	0.00	0.25	0.75	1.0	1.515
	Total school					7.0	0.684
	Sleeping	1.00	0.00	0.00	0.00	10.0	0.291
	Weighted daily average						0.843

<sup>a</sup>. The infant and child represent the same scenario.

<sup>b</sup>. The student matures to the age of 18 during his exposure. The weighted daily average breathing rate (m<sup>3</sup> d<sup>-1</sup>) for age 9–18 are as follows: age 10, 0.732; age 12, 0.910; age 14, 1.00; age 16, 1.19; and age 18, 1.26.

## Uncertainty Factors for Annual-Average Dispersion Estimates

We accounted for uncertainty in atmospheric transport modeling for continuous releases by three multiplicative stochastic correction factors that accounted for uncertainty in the dispersion estimate, the meteorology, and deposition and plume depletion ([Rood 1999a](#); [Rood and Grogan](#)



1999b). Dispersion uncertainty was based on distributions of predicted-to-observed ratios from field tracer experiments using the Gaussian plume and other models, including RATCHET. These values were derived from literature reviews and results from studies specific to this project. Meteorological uncertainty arises because we used 5 years of meteorological data spanning a recent time period (1989–1993) to define an annual average  $\Pi/Q$  value that was applied to the specific years of the assessment period (1953–1989). This correction factor was derived from studies performed for the Fernald Dosimetry Reconstruction Project ([Killough et al. 1998](#)) and additional site-specific comparisons made at Rocky Flats. Deposition and plume depletion uncertainty factors were calculated using the Monte Carlo sampling features of RATCHET. All correction factors were distributed lognormally and were combined with the source term uncertainty to yield distributions of predicted concentrations at selected receptor locations.

The derivation and rationale behind the distributions assigned to each uncertainty correction factor are described in [Rood \(1999a\)](#) and [Rood and Grogan \(1999b\)](#) and summarized in Table 14. The three correction factors are independent of one another. The dispersion correction factor is assumed to be correlated from year to year (correlation coefficient = 1.0) and, therefore, is sampled once during a Monte Carlo realization. The other correction factors are independent from year to year and are sampled  $n$  times during a Monte Carlo realization, where  $n$  is the number of exposure years in the entire Monte Carlo simulation. Integration of these correction factors into the *TIC* estimates is discussed in the [“Mathematical Model”](#) section of this report.

**Table 14. Summary of Uncertainty Correction Factors Applied to Annual Average Air Concentration Predictions**

Receptor distance (km)	Dispersion uncertainty		Meteorology uncertainty		Depletion uncertainty	
	GM <sup>a,b</sup>	GSD <sup>c</sup>	GM	GSD	GM	GSD
<4	1.1	2.2	1.0	1.7	1.0	1.05
8	1.1	2.0	1.0	1.7	1.0	1.09
12	1.1	2.0	1.0	1.7	1.0	1.12
16	1.1	2.0	1.0	1.7	1.0	1.14
20	1.0	2.2	1.0	1.7	1.0	1.16
24	1.0	2.2	1.0	1.7	1.0	1.17
28	1.0	2.2	1.0	1.7	1.0	1.18
>32	1.0	2.2	1.0	1.7	1.0	1.18

<sup>a</sup> GM = geometric mean.

<sup>b</sup> Dispersion uncertainty GM is the inverse of the GM of predicted-to-observed ratios.

<sup>c</sup> GSD = geometric standard deviation.

## MODEL VALIDATION

Model validation is the process where model predictions are evaluated against an independent set of measurements of like quantities. Model validation is the means by which the accuracy of the model is evaluated. Accuracy is a measure of how close the model prediction is to a similar *measured* quantity. We contrast this with model precision, which is ability of the model to reproduce the same result upon repeated trials, given uncertainty in the input parameters. The parameter uncertainty analysis discussed earlier was used to establish model precision.

In environmental dose reconstruction, model-predicted and -measured quantities are typically concentrations of contaminants in environmental media. It is important that the environmental data sets used in model validation exercises are independent of data sets used to develop and calibrate the model. Therefore, it is not valid to compare predicted concentrations with measured values that were originally used to calibrate the model. For example, the source term for the 903 Area was developed and calibrated using measurement data from the S-8 sampler. Therefore, comparisons of model predicted air concentrations with S-8 sampler data do not provide evidence of model validation. It is also important that the measured quantities selected for comparison are within the same time and spatial resolution as the model. Spatial resolution of the model was 1 km and temporal resolution was typically 1 year. However, provisions were made in the code to provide concentrations averaged over the duration of a discrete event (9–26 hours).

Environmental monitoring data potentially useful for model validation have been compiled and evaluated in [Rope et al. \(1999\)](#). Environmental measurements include plutonium concentrations in soil, vegetation, ambient air, water, and lake sediment. Other measurements include plutonium deposition flux and miscellaneous plutonium measurements in lichens, human tissue, human urine, and cattle. The model described in the previous sections provides estimates of concentrations in ambient air and soil. Additional modeling and assumptions were necessary to predict concentrations in other environmental media. In general, we limited our comparisons to the environmental media of soil, ambient air, lake sediment, and vegetation.

The model comparisons presented in this section are qualitative. That is, we did not do any statistical analysis between the predicted and observed values or base the performance of the model on some numeric criteria. This section provides information to evaluate the model predictions in terms of measurable quantities in the environment.

### Model Comparisons with Plutonium Soil Data

This section presents comparisons of model-predicted plutonium concentrations in soil with measured values. Soil represents a significant sink for plutonium deposited from the air. Plutonium is relatively insoluble and immobile under most conditions, thereby allowing it to accumulate in the upper soil layers. These comparisons are useful for validating the total plutonium released from the site, and in particular, releases from the 903 Area because the 903 Area is believed to have been responsible for most of the offsite plutonium contamination from Rocky Flats releases.

Soil measurement data are reviewed and evaluated in Chapter 8 of [Rope et al. \(1999\)](#). Few measurements exist for years before 1969. What information has been uncovered refers to total alpha activity in soils from scattered locations. The earliest studies ([Thackeray 1953](#)) were

designed to detect alpha activity trends so that any subsequent RFP-caused contamination would be detected. This sampling program also included vegetation, water, and algae. Additional sampling was performed in September and November 1957 following the September glove box fire in Building 771. The narrative in the September report ([Hammond 1957](#)) stated that offsite soil and vegetation samples indicated possible low-level contamination from the fire. However, offsite background soil measurements were not taken to compare with 1957 postfire data. [Rope et al. \(1999\)](#) concludes that it is essentially impossible to draw specific conclusions from these data.

Several sampling studies were initiated following the Building 776/777 glove box fire in May 1969 ([CCEI 1970](#); [Krey and Hardy 1970](#); [Poet and Martell 1972](#)). These studies showed a well-defined plume of plutonium extending eastward from the RFP. The plutonium plume in soil was attributed to releases from the 903 barrel storage area. Numerous other studies of plutonium in offsite soil performed by various agencies followed the initial reports of offsite contamination and continue to this day. These studies are reviewed in [Rope et al. \(1999\)](#).

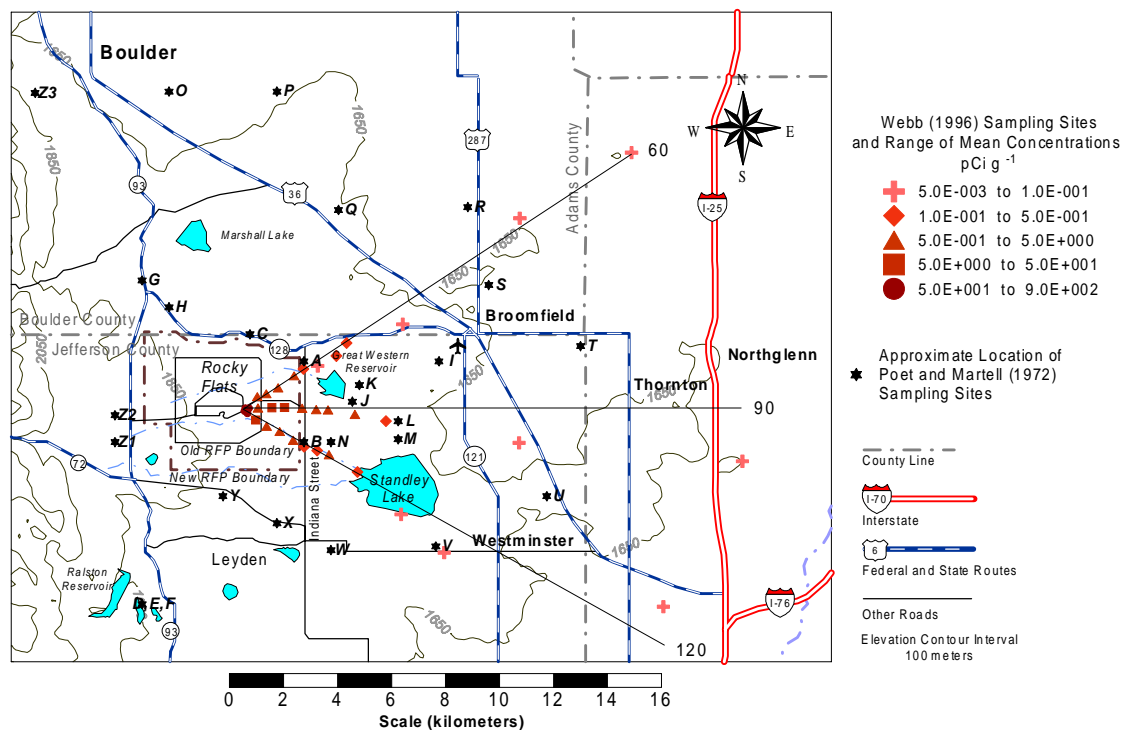
In reviewing these data for model validation purposes, it became apparent that there were several technical problems in comparing these data to model predictions. First, the sampling depth and protocol were not consistent. Sampling depth used in the studies varied from several millimeters to ~20 cm. The sampling depth and fraction of the total deposited activity in that depth must be known for a valid comparison to be made between predicted and measured concentrations. Second, most of these studies, with the exception of [Webb \(1996\)](#), did not systematically measure plutonium in soil with distance and direction from the 903 Area. Systematic sampling is important if we are to evaluate the spatial trend of plutonium concentrations in soil. The [Webb \(1996\)](#) study measured plutonium in soil along four transects out to distances of ~20 km. A uniform sampling protocol was used for all samples and consideration was given to soil density, rock volume, and distribution of plutonium with depth. [Webb \(1996\)](#) also provided inventory estimates with uncertainty within his study domain.

We concluded that the [Webb \(1996\)](#) data set was the best source of soil model validation data available to us. We reviewed larger data sets, such as those compiled by [Litaor et al. \(1994\)](#). However, sampling depth and the year of sampling was inconsistent, making it difficult to compare model predictions with measured data. We also used a second data set reported in [Poet and Martell \(1972\)](#) in the comparison. These measurements were made in late 1969 and early 1970 and, therefore, provide a comparison of the modeled temporal depth distribution. Unfortunately, Poet and Martell did not sample systematically with distance and direction from the 903 Area. However, a uniform sampling protocol was adhered to for all samples.

We acknowledge that we also used portions of the [Webb \(1996\)](#) and [Poet and Martell \(1972\)](#) data set to develop plutonium depth distributions. Consequently, these data sets are not entirely independent. Nevertheless, uncertainties associated with the source term, transport, and deposition of plutonium in airborne plumes far outweigh the uncertainties associated with the depth distribution. Comparisons of plutonium soil inventories however, are completely independent (because depth distribution is not considered) and provide perhaps the best validation of total plutonium quantities released from the RFP. A description of the [Webb \(1996\)](#) and [Poet and Martell \(1972\)](#) data sets follows.

### The [Webb](#) (1996) Data Set

Webb measured plutonium in soil along four transects that followed azimuths of 60°, 90°, 120°, and 150° true bearing and extended out to approximately 20 km from the origin at the 903 Area. Samples taken along the 150° transect were near the background concentration of 0.029–0.12 pCi g<sup>-1</sup> (1.1 to 4.6 Bq kg<sup>-1</sup>) measured by Webb and, therefore, were not included in the analysis ([Figure 15](#) and [Table 15](#)). Sampling sites were selected in natural, undisturbed areas where possible. Six to thirteen 100-m<sup>2</sup> macroplots were spaced at exponentially increasing distances from the 903 Area. Of particular interest in the evaluation was the spatial variance in the plutonium data to enable quantification of uncertainty in the results. To accomplish this, samples were replicated at four, randomly located 1-m<sup>2</sup> microplots within each macroplot.



**Figure 15.** Location of measurement sites from [Webb](#) (1996) and [Poet and Martell](#) (1972). The mean concentration in the 0–3-cm layer from the Webb data set is shown. The 150° transect is not shown and was not used in Webb’s analysis. Locations of the Poet and Martell sites were approximated from Figure 1 in [Poet and Martell](#) (1972).

The general sampling procedure used by [Webb](#) (1996) follows. (1) Clip the standing vegetation at ground level inside a 1250 cm<sup>3</sup> frame then scrape the 3 mm of surface soil using the Colorado Department of Health scoop and template method ([CDH](#) 1977). (2) Clip the standing vegetation at ground level inside a 625 cm<sup>2</sup> frame then excavate a 25 × 10-cm area of soil in 3-cm layers to a depth of 21 cm. (3) Clip standing vegetation at ground level inside a 625 cm<sup>2</sup> frame then excavate a 25 × 15 cm area of soil to a depth of 3 cm for three of the four microplots at all offsite macroplot locations. (4) Clip the standing vegetation at ground level inside a 625 cm<sup>2</sup> frame then excavate a 5 × 5-cm area of soil to a depth of 21 cm. (5) Collect site information

including location, vegetation cover, and rock quantity. The rock volume was measured for each site by removing the material from a ~13-L hole in front of the area that was sampled and sieving it through a screen with a 5-mm square mesh. The collected rocks were then weighed and converted to volume using an estimated density of  $2.65 \text{ g cm}^{-3}$  which is the density of quartz. Ten background sites were also measured along the front range of Colorado in area only affected by global fallout using the same soil sampling procedure. Soil density was also measured at each of the sampling sites. This density represents density of the soil excluding the rocks.

**Table 15. Plutonium Concentrations Measured by [Webb](#) (1996) in the 0–3 cm Soil Layer**

Macroplot location code	Azimuth	Distance from 903 Area (km)	Number of		Standard deviation (pCi g <sup>-1</sup> )
			0–3 cm samples	Mean (pCi g <sup>-1</sup> )	
AX1	90	0.28	3	892	222.
AX2	90	0.41	5	193	63.2
AX3	89	0.74	6	2.14	1.85
AX4	89	1.26	4	15.8	4.73
AX5	89	1.73	4	6.94	1.55
AX6	90	2.39	4	3.11	0.865
DX1	91	2.9	4	2.59	0.581
DX2	91	3.34	5	0.892	0.614
DX3	93	4.35	4	0.784	0.368
DX4	95	5.51	4	0.198	0.03541
DX5	97	10.5	4	0.0646	0.0111
DX6	96	18.8	4	0.0454	0.0138
BX1	59	0.84	4	4.08	0.899
BX2	63	1.18	4	3.99	1.05
BX3	63	1.67	4	0.670	0.435
BX4	60	2.41	4	0.611	0.565
BX5	59	2.83	4	0.191	0.124
EX1	62	3.35	5	0.0508	0.00649
EX2	62	4.13	4	0.241	0.0714
EX3	59	4.7	2	0.266	0.211
EX4	63	6.86	5	0.0800	0.0784
EX5	56	12.6	4	0.0259	0.0143
EX6	57	17.4	4	0.0951	0.0251
CX1	112	0.27	4	150.	33.2
CX2	117	0.4	4	50.8	18.2
CX3	123	0.79	4	12.3	10.1
CX4	122	1.26	4	1.47	0.183
CX5	119	1.77	4	3.16	1.05
CX6	120	2.37	3	1.41	0.300
CX7	120	2.84	4	0.124	0.0570
FX1	118	3.33	4	0.497	0.128
FX2	117	3.79	4	0.805	0.384
FX3	118	5.02	4	0.207	0.146
FX4	123	7.22	5	0.0270	0.0124
FX5	125	9.33	5	0.0340	0.0168
FX6	115	17.4	4	0.00541	0.00270
Background			10	0.058	0.021
Community			11	0.061	0.029

Samples were prepared by dry sieving to <2 mm, then homogenized, ground, split, and packaged for analysis. Plutonium analysis used a batch-leaching procedure developed to accommodate soil samples up to 50 g. A passive ion-implanted surface barrier detector was used to measure plutonium activity using alpha spectroscopy.

Approximately 1400 individual plutonium measurements were made during the project. Plutonium concentrations in soil were found to decrease rapidly with depth into the soil and distance from the 903 Area. Concentrations in the 0–3-cm layer for offsite locations ranged from background ( $\sim 0.05$  pCi g<sup>-1</sup> [2 Bq kg<sup>-1</sup>]) to  $\sim 3$  pCi g<sup>-1</sup> (120 Bq kg<sup>-1</sup>). Concentrations along the 90° transect were elevated relative to the other transects.

The sample preparation procedure was an important aspect of this study. Webb argued that the rocks do not contain significant quantities of plutonium and, therefore, must be considered when making an inventory estimate. He attributed much of the difference between his inventory estimates and those of other researchers to his consideration of rock volume.

### The [Poet and Martell](#) (1972) Data Set

[Poet and Martell](#) (1972) collected 30 samples in the 0–1-cm soil layer at 28 locations around the RFP ([Figure 15](#) and [Table 16](#)). Depth profiles were measured at three of these locations. At two sampling sites (B and C), replicate samples were taken. The average values among the replicates were used in the model comparison. Background concentrations were measured at seven additional sites located in Derby, northeast Denver, Aurora, east Denver, southeast Denver, southwest Denver, and Golden. The mean concentration measured at these sites was 0.036 pCi g<sup>-1</sup>, with a standard deviation of 0.01 pCi g<sup>-1</sup>. This value was within the range of background measured by [Webb](#) (1996) of 0.029–0.12 pCi g<sup>-1</sup>. All samples were collected from even, undisturbed sites. The vegetation was cropped closely and discarded. Each soil sample was collected over an area of about 1000 cm<sup>2</sup> by spatula, with special care to provide samples of the stated depth as uniformly as possible. Soil samples were air dried, pulverized to breakdown soil aggregates, sieved to remove particles >0.05-cm diameter, and thoroughly mixed. The measured density of the screened and dried soil samples was about 1.0 g cm<sup>-3</sup>.

[Poet and Martell](#) (1972) identified locations of 28 sampling sites on a map. No other information, such as the latitude and longitude or universal transverse mercator (UTM) coordinates of the sample sites, was provided. The location of each sampling site was “eyeballed” on to the project base map ([Figure 3](#)), and UTM coordinates were defined. Predicted concentrations were then obtained at the model node nearest to each of the sampling sites. This value was then compared to the corresponding measured data.

### Soil Concentration Comparisons with [Webb](#) (1996) Data

We extracted predicted concentrations in the 0–3-cm soil layer for 1989 from the model output by interpolating concentrations between each of the output nodes along each of Webb’s transect lines. The model does not do computations beyond 1989; however, soil concentrations in 1989 are essentially the same as would be predicted for 1994, the year the sampling was performed. The nearest node was located about 500 m east of the 903 Area. Consequently,



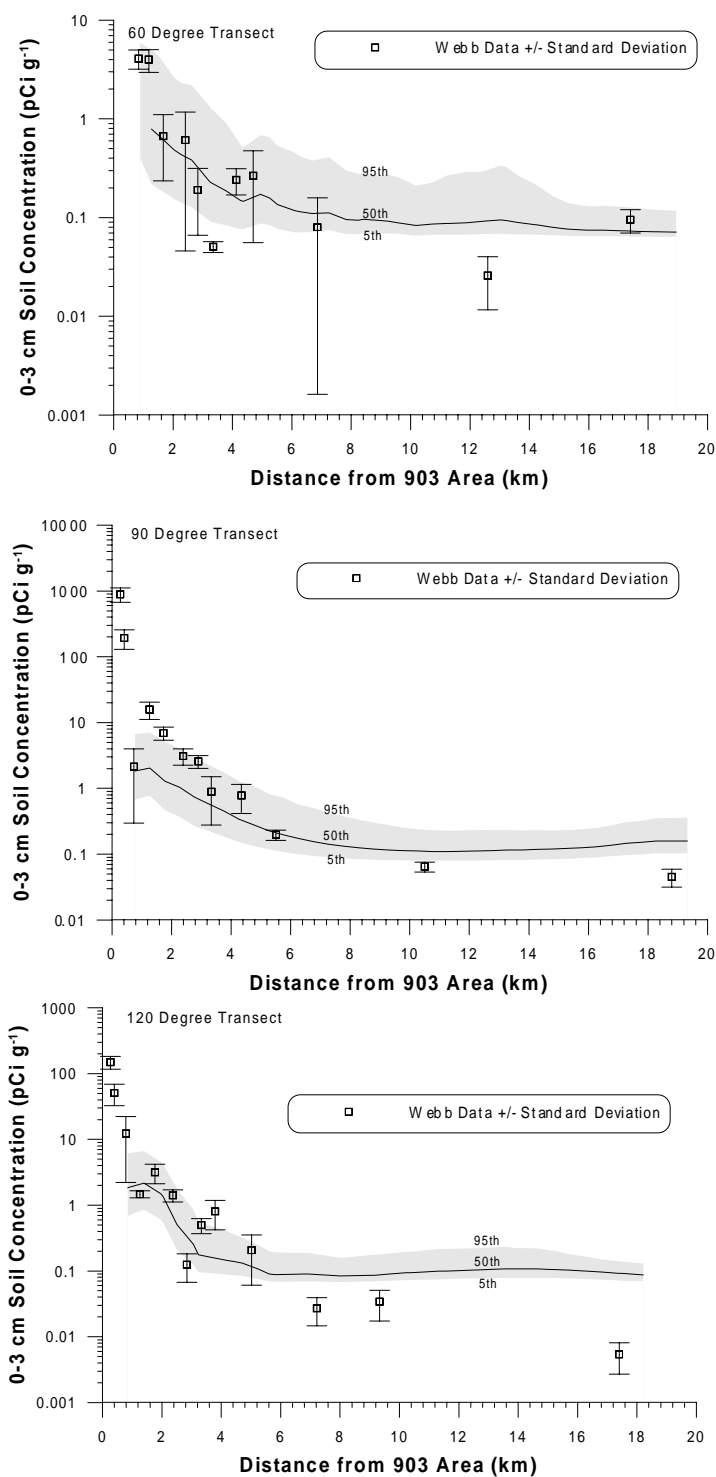
**Table 16. Plutonium Concentrations Measured by [Poet and Martell](#) (1972) in the 0–1 cm Soil Layer**

Location	UTM E <sup>a</sup>	UTM N <sup>a</sup>	Concentration (pCi g <sup>-1</sup> )	Location	UTM E <sup>a</sup>	UTM N <sup>a</sup>	Concentration (pCi g <sup>-1</sup> )
A	485850	4417050	0.581	N	486850	4414050	0.603
B	485850	4414050	1.05	O	480850	4427050	0.0306
B	485850	4414050	6.08	P	484850	4427050	0.0486
C	483850	4418050	0.0414	Q	487129	4422660	0.018
C	483850	4418050	0.0257	R	491923	4422770	0.0351
D	479850	4408050	0.0576	S	492696	4419880	0.0459
E	479850	4408050	0.126	T	496099	4417610	0.0788
F	479850	4408050	0.0941	U	494850	4412050	0.144
G	479850	4420050	0.141	V	490735	4410190	0.244
H	480850	4419050	0.0581	W	486850	4410050	0.0149
I	490850	4417050	0.774	X	484850	4411050	0.0842
J	487642	4415550	0.518	Y	482850	4412050	0.189
K	487901	4416170	0.396	Z1	478850	4414050	0.0527
L	489343	4414830	0.0522	Z2	478850	4415050	0.108
M	489343	4414160	0.167	Z3	475888	4427000	0.137

<sup>a</sup>UTM coordinates were approximated in this study.

predicted concentrations within 500 m of the 903 Area were unavailable. Predicted results were extracted separately for the 5th, 50th, and 95th percentile values and are shown in [Figure 16](#) along with the corresponding measured data. We added the mean background value determined by [Webb](#) (1996) of 0.06 pCi g<sup>-1</sup> to all predicted values. The measured data include plutonium from RFP and global fallout sources. The error bars shown in [Figure 16](#) on Webb's measurements represent the standard deviation of the sample replicates taken within each macroplot. Predicted and measured concentrations decrease as a function of distance from the 903 Area. A slight increase in the predicted soil concentration is observed in all transects around 10 to 16 km east of the 903 Area. For the 60° transect, the increase is attributed to the higher roughness coefficient assigned to the city of Broomfield (0.6 m), compared to the surrounding area (0.07 m). Higher roughness coefficients yield higher deposition velocities. Deposition from the 1957 fire plume is responsible for the increase observed for the 90° and 120° transect, along with the higher roughness coefficient for the city of Westminster in the 120° transect.

In general, the model underpredicted concentrations close to the 903 Area and out to the Rocky Flats property boundary at Indiana Street (~2 km west of the 903 Area). Beyond Indiana Street, model predictions are more inline with measured data. Underprediction within the plant buffer zone (west of Indiana Street) is not surprising because the predicted 903 Area releases did not include particles >30 µm. Activity transported from the 903 Area because of saltation effects were also not included in the model. Plutonium attached to larger soil particles would tend to deposit quickly after being suspended, resulting in high plutonium concentrations near the 903 Area.



**Figure 16.** Predicted and measured soil concentrations in the 0–3-cm layer for the three transects originating from the 903 Area and extending eastward. Predicted results include 0.06 pCi g<sup>-1</sup> from background sources. Measured data are from [Webb](#) (1996) and represent plutonium from Rocky Flats sources and that from global fallout.



The measured data reflect the rather large variability of plutonium soil concentrations in the model domain. This variability can be attributed to many different factors, including local geochemical and geologic conditions, vegetative cover, and human and animal disturbances.

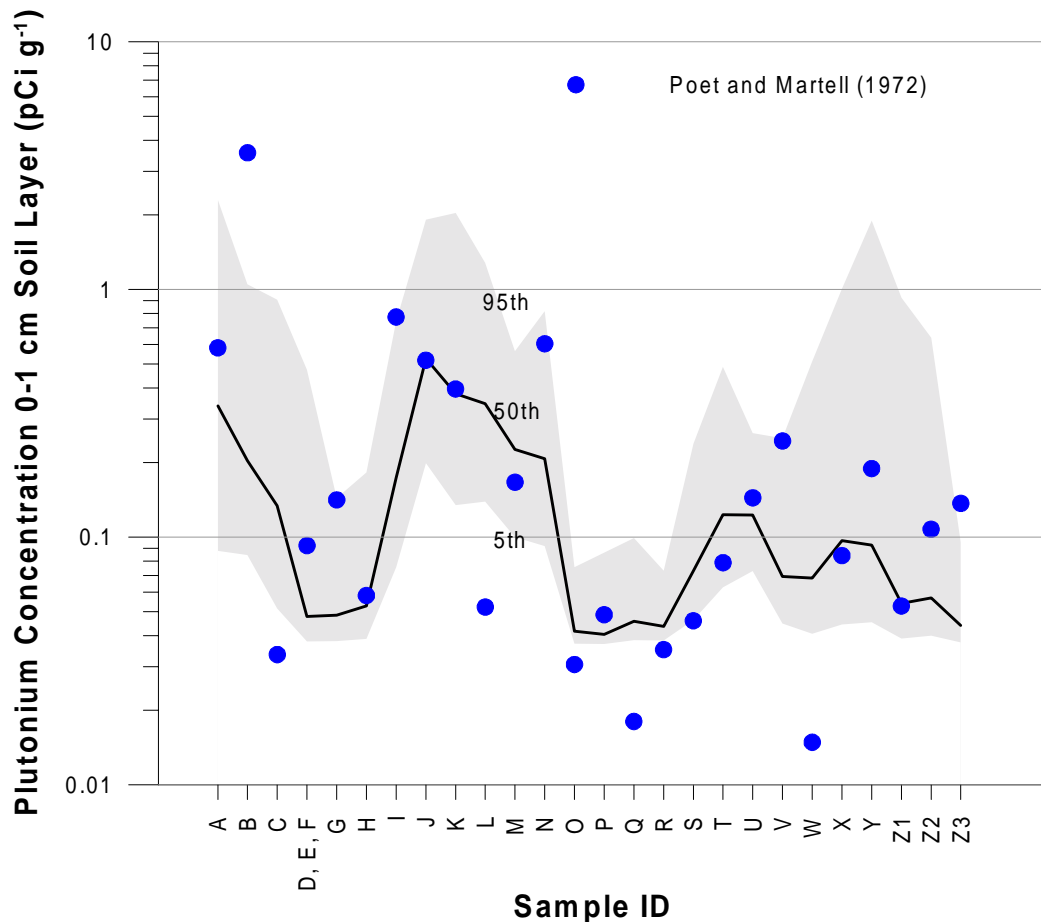
Qualitatively, there appears to be reasonable agreement between the model-predicted values and soil concentrations measured by [Webb](#) (1996) at distances >2 km. Discrepancies occurred at locations close to the 903 Area (<2 km), where deposition of large, nonrespirable particles is suspected to have occurred. At distances >8 km, measured soil concentrations approach background levels from global fallout. When considering the background plutonium levels in soil, model-predicted soil concentrations appear to be overestimated at distances >8 km. These observations suggest the fraction of activity attached to the larger particles may have been underestimated. Larger particles in model simulations would have resulted in greater deposition near the source and lower deposition at greater distances. Therefore, predicted plutonium intake and risk may be biased high for 903 Area releases because larger particles (>15  $\mu\text{m}$ ) are not respirable.

### Soil Concentration Comparisons with [Poet and Martell](#) (1972) Data

Predicted and measured soil concentrations at the [Poet and Martell](#) (1972) sampling sites are shown in [Figure 17](#). Concentrations were predicted for 1970, the year the sampling was performed. The mean background concentration reported in [Poet and Martell](#) (1972) of 0.039 pCi  $\text{g}^{-1}$  was added to all predicted concentrations. Predicted values represent the concentration in the 0–3-cm layer. Because we assumed homogenous mixing in the 0–3-cm layer, the *predicted concentration* in the 0–1-cm layer would be the same. Sampling sites were arranged in alphabetical order and are not intended to show any spatial trend. Two of the 26 measurements were greater than the corresponding predicted values at the 95% level. The model substantially underpredicted plutonium concentrations at sampling site B, which lies on the 120° transect line near Indiana Street. Other samples taken near this transect (N and V) were also near the 95th percentile value, which suggests a tendency of the model to underpredict concentrations along this transect. At six of the sampling sites, the model-predicted concentrations at the 5% level were greater than the measured concentrations.

### Soil Inventory Comparisons

Inventory refers to the total amount of plutonium in the soil column in a defined area. Predicted plutonium soil inventories were compared with inventory estimates made by Webb and other researchers and reported in [Webb](#) (1996). The inventory estimates were based on extrapolation of measured plutonium concentrations in soil. Inventories were reported for the Webb study area and for the total area affected by Rocky Flats plutonium. The Webb study area encompassed an area of 209  $\text{km}^2$  and was bounded by the 60° and 120° transects out to a distance of about 20 km from the 903 Area. The total area affected by Rocky Flats plutonium varied among researchers, but it was estimated to range from 600 to >2000  $\text{km}^2$ . Our calculations assumed the entire model domain (2200  $\text{km}^2$ ) as the total affected area. However, most of the plutonium in soil was restricted to an area of about 1000  $\text{km}^2$  where deposition from the 903 Area releases is postulated to have occurred.



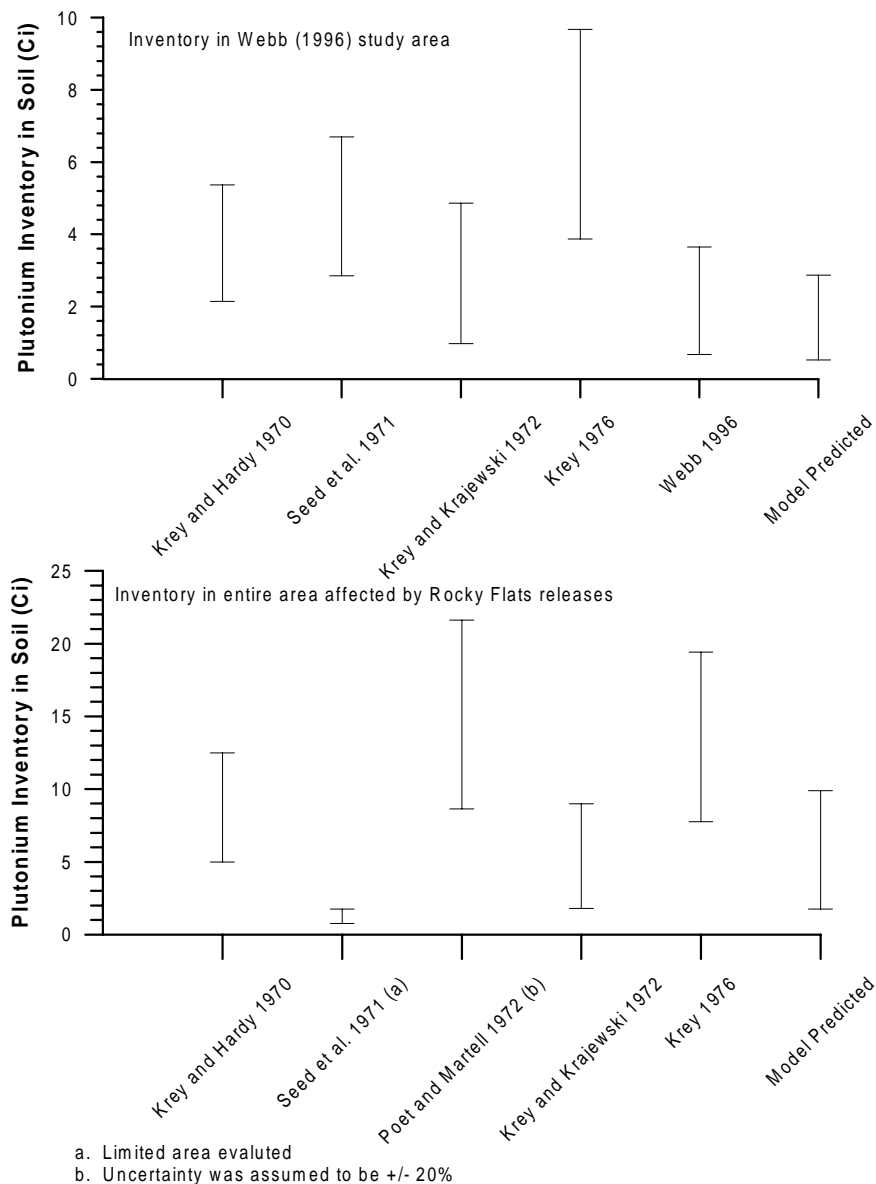
**Figure 17.** Comparison of model-predicted soil concentrations and measured concentrations by [Poet and Martell](#) (1972). The shaded area represents the 5th and 95th percentile range of predicted concentrations. Sampling locations were not arranged to show any spatial trend.

The empirical equations developed by Webb were not extrapolated to regions beyond his study area. Therefore, inventory estimates made by Webb were restricted to only that area bounded by his study area. Model-predicted inventories for the Webb study area were generated by integrating predicted surface concentrations over an area bounded by the 120° and 60° transects.

Contributions from background were subtracted by Webb so that the inventory estimates represented only Rocky Flats plutonium sources. Where possible, Webb provided uncertainty bounds on estimates made by other researchers. The uncertainty calculated for his own inventory estimates was based on Monte Carlo sampling of the regression equations used to predict plutonium in soil as a function of distance and direction from the 903 Area.

Webb's inventory estimates within his study area were generally lower than those made by other researchers. He attributed this outcome to the exclusion of rock volume in estimating inventories and other factors including depth distributions, soil density, and contributions from fallout plutonium. Comparisons of inventory estimates are shown in [Figure 18](#). Model-predicted plutonium inventories in the Webb study area (upper graph in [Figure 18](#)) were slightly less than

those made by Webb, but the two were in general agreement. We attribute some of our model's underprediction to its underestimation of plutonium soil concentrations near the 903 Area.



**Figure 18.** Plutonium soil inventories estimated from soil concentration measurements and model predictions. The upper graph represents the inventory in the 209 km<sup>2</sup> Webb study area. The lower graph represents the entire area affected by Rocky Flats releases. Inventory estimates were obtained from Table 2-2 in [Webb](#) (1996). Predicted inventory values are the 5th and 95th percentiles.

Underestimation of plutonium in soil near the 903 Area was attributed to exclusion of large particles (>50 µm) from the 903 Area source term. Most of the other researchers had inventory

estimates that overlapped the uncertainty bounds of the model-predicted inventories. Only [Krey \(1976\)](#) shows a substantially higher inventory estimate than the other researchers.

Plutonium inventories in the entire model domain (lower graph in [Figure 18](#)) shows the model-predicted values are in generally good agreement with the other researcher's estimates. Plutonium inventory estimates made by [Poet and Martell \(1972\)](#) are substantially greater than those of the other researchers. The higher inventory estimate reported in [Poet and Martell \(1972\)](#) was based on the measured ratio of  $^{239}\text{Pu}$  to  $^{90}\text{Sr}$  in samples collected 48–65 km northeast of the RFP. Using the  $^{90}\text{Sr}/^{239}\text{Pu}$  ratios, they proportioned a fraction of the plutonium measured at distant locations to RFP sources and integrated over that area to obtain their inventory estimate.

Overall, the model-predicted plutonium inventories in soil are within about a factor of 2 of the inventory estimates made by extrapolating plutonium soil concentration data. These results provide some validation of the total quantities of plutonium released from the plant. However, most of the plutonium in soil in the model domain is believed to have originated from the 903 Area. Contributions to the total plutonium soil inventory in the model domain from the 1957 fire event were between 0.44 Ci (5th percentile) and 2.6 Ci. (95th percentile) based on model simulations. Model-predicted soil concentrations from routine operations and the 1969 fire would be difficult to discern from background plutonium soil concentrations. While the 1957 fire was estimated to be the largest offsite release of plutonium from the plant (median estimate of ~20 Ci, [Voillequé 1999b](#), it deposited far less plutonium to the soil in the model domain than 903 Area releases (median release estimate of ~3 Ci, [Weber et al. 1999](#)). We attribute this observation to (a) particles sizes for the 1957 releases were smaller than 903 Area releases, resulting in lower gravitational settling velocities and deposition, (b) winds were relatively light during the 1957 fire compared to the discrete 903 Area releases and resulted in lower calculated deposition velocities, and (c) 1957 fire releases were from an elevated plume compared to ground-level releases for 903 Area.

### Model Comparisons with Ambient Air Monitoring

This section compares model-predicted plutonium concentrations in ambient air to corresponding measured values. Ambient air monitoring is the monitoring of air in the open environment; it is different from effluent air monitoring, which monitors effluent air leaving facilities via stacks, exhaust fans, etc. We found air monitoring data were useful (for reasons discussed later in the section) for validating resuspension of contaminated soil in the 1970s and 1980s and releases from the 1969 glove box fire in Building 776. Air monitoring data were also used to calibrate 903 Area releases from 1964–1969.

### Review of Relevant Air Monitoring Data

Air monitoring data taken during the Rocky Flats operational period are reviewed, analyzed, and interpreted in [Rope et al. \(1999\)](#). Air monitoring data include measurements performed by the RFP contractor (contractor monitoring) and monitoring performed by independent agencies. Independent agencies included the CDPHE, DOE's Health and Safety Laboratory (HASL), the EPA, and the U.S. Public Health Service. The HASL is now called the Environmental Measurements Laboratory. Operated by the DOE and its predecessors (Energy Research and Development and the Atomic Energy Commission), the HASL was not independent of the DOE complex, but it was not involved in operating the RFP. In 1973, the U.S. Public Health Service

radiological air monitoring program was integrated into the Environmental Radiation Ambient Monitoring System of the EPA Office of Radiation Programs. The U.S. Public Health Service began monitoring plutonium in airborne particulates in Denver air in 1965.

The RFP contractor began onsite ambient air monitoring at a single station in 1952. By early 1953, 10 onsite stations had been established; in 1969, two additional stations were added. The routine air monitoring equipment were low volume air samplers that operated at a flow rate of  $0.057 \text{ m}^3 \text{ min}^{-1}$ . Portable high-volume samplers were maintained for accident events like the fire in 1957 and operated at a flow rate of 0.34 to  $1.1 \text{ m}^3 \text{ min}^{-1}$ . Onsite sampler filters were typically changed daily, around 8:15 a.m. The filter changes for offsite samplers were made weekly in the late 1960s; however, the samplers were programmed to operate intermittently for 5 minutes per hour. The volume of air sampled in a week was consequently less than the onsite samplers obtained in a day ([Rope et al. 1999](#)).

The overall quality of the air monitoring data during the 1950s are assessed in [Rope et al. \(1999\)](#). In the 1950s (particularly 1955–1960), 4-hour gross alpha counts were made. That is, the count of the air sample filter was made 4 hours after collection and included large contributions from natural alpha-emitting radionuclides like radon decay products. Consequently, the 4-hour count results are of no value in assessing the concentrations of long-lived alpha emitters like plutonium released from Rocky Flats.

In the 1960s, the RFP contractor began measuring total long-lived alpha (TLL $\alpha$ ) activity in air, in contrast to the 4-hour count. Filters were analyzed 1 week after collection to allow time for short-lived alpha activity to decay before counting the sample. However, the detection limits during the 1960s were still relatively high, resulting in many of the samples being at or below the minimum detectable concentration (MDC) of 0.21 counts per minute (cpm) or  $5.5 \text{ fCi m}^{-3}$  ([Rope et al. 1999](#)). For example, for 12 onsite samplers from October 1964 through December 1971, 45 to 80% of the daily measurements were less than the MDC for total long-lived alpha activity. The sampler with the fewest number of results less than the MDC was S-8. This sampler was located near the 903 Area and was strongly influenced by suspension of contaminated soil during the 1960s. This sampler was used to calibrate 903 Area releases ([Weber et al. 1999](#)).

Data from community samplers that were maintained by the site contractor during the 1960s were also obtained. Original records were obtained for the years 1966–1971. [Rope et al. \(1999\)](#) states that the sampling and analysis technique for TLL $\alpha$  activity in these samplers suffered from poor sensitivity because of short count times, small volumes of air sampled, and variable counter background. In addition, long-lived alpha activity from naturally occurring radionuclides collected on the air filters was significant and could not be distinguished from Rocky Flats materials. [Rope et al. \(1999\)](#) concludes that quantitative use of the contractor offsite monitoring data before 1970 for this historical public exposures study was not warranted.

In the 1970s, plutonium-specific analysis of routine air monitoring stations maintained by the RFP contractor began. As of 1990, there were 51 samplers in the ambient air sampling network. Twenty-three samplers were located at the RFP industrial site, 14 were located along the boundary bordered by major highways, and 14 were placed in community locations. Detection limits also improved during this time from  $0.01 \text{ fCi m}^{-3}$  in the mid-1970s to  $\sim 0.002 \text{ fCi m}^{-3}$  by 1986 ([Rope et al. 1999](#)).

Monitoring of plutonium in air near Rocky Flats by agencies other than the Rocky Flats contractor started in 1965, with the U.S. Public Health Service monitoring in Denver and other U.S. cities and HASL monitoring locations around the world ([Rope et al. 1999](#)). Measurements

were also made by the HASL at locations near the 903 Area at Rocky Flats and three other nearby stations between 1970 and 1981. The Colorado Department of Health began monitoring plutonium in airborne particulates at a station near the 903 Area in 1969; other onsite and offsite stations were added in the early 1970s ([Rope et al. 1999](#)).

Based on the limitations of the monitoring data discussed above, we restricted our comparisons of predicted and measured air concentrations to those data taken after 1970 because data before that time were either nonexistent or suffered from poor detection limits and/or significant interference with other naturally occurring radionuclides. In addition, we limited our comparisons of predicted and measured plutonium air concentrations to locations outside the RFP industrial area, with one exception as noted in the next paragraph. Model resolution within the industrial area is poor (1-km grid node spacing), making it impossible to distinguish between samplers located several hundred meters apart. In addition, air concentrations within the industrial area are influenced by building wakes and small, local sources of plutonium contamination (such as the solar evaporation ponds). Building wake effects and releases from localized sources of plutonium contamination onsite were not included in the model. These processes are important for predicting exposures to people working in the industrial area; however, their overall impact on air concentrations offsite is small ([Rood 1999b](#); [Rood and Grogan 1999a, 1999b](#)). Because the primary purpose of this study is to estimate offsite exposures to the public, we did not include these processes in the model and restricted our comparisons to air samplers located offsite.

An exception to the limitation stated in the preceding paragraph was made for air monitoring data taken onsite during the May 11, 1969, glove box fire in Building 776. Community air monitoring stations recorded concentrations at or near fallout background or less than detection limits during the fire. Therefore, model comparisons with these data could not be performed. Contractor-operated onsite stations, however, did measure concentrations greater than background and above the MDC during this time. Therefore, these data were used because they were the only source of local air monitoring data for comparison with model predictions.

### **Model Comparisons with Post 1970 Data**

Four monitoring stations were selected for evaluating the annual average plutonium concentrations in air, and they include measurements made by the RFP contractor, HASL, and CDPHE. The four locations were (1) the old RFP boundary where it intersects with the east access road, (2) Indiana Street where it intersects with the east access road, (3) the City of Broomfield, and (4) Leyden. Samplers maintained by HASL were located at the old RFP boundary (HASL 4) and near the intersection of Indiana Street and the east access road (HASL 2). The RFP contractor stations included samplers S-32/37<sup>c</sup> located near the HASL 2 sampler and stations in Broomfield and Leyden. The CDPHE sampler D-5 was also located on Indiana Street, but it was located about 500 m south from the HASL and contractor stations. Annual average measured concentrations at each location were determined by [Rope et al. \(1999\)](#). Annual contributions from fallout sources were estimated from U.S. Public Health Service monitoring of air in Denver and were subtracted from the annual average measured concentrations to yield net

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<sup>c</sup> S-32 was the sampler designation before 1975. After 1975, the sampler was designated as S-37.



annual average plutonium concentrations. The net plutonium concentrations were compared to predicted concentrations from Rocky Flats releases.

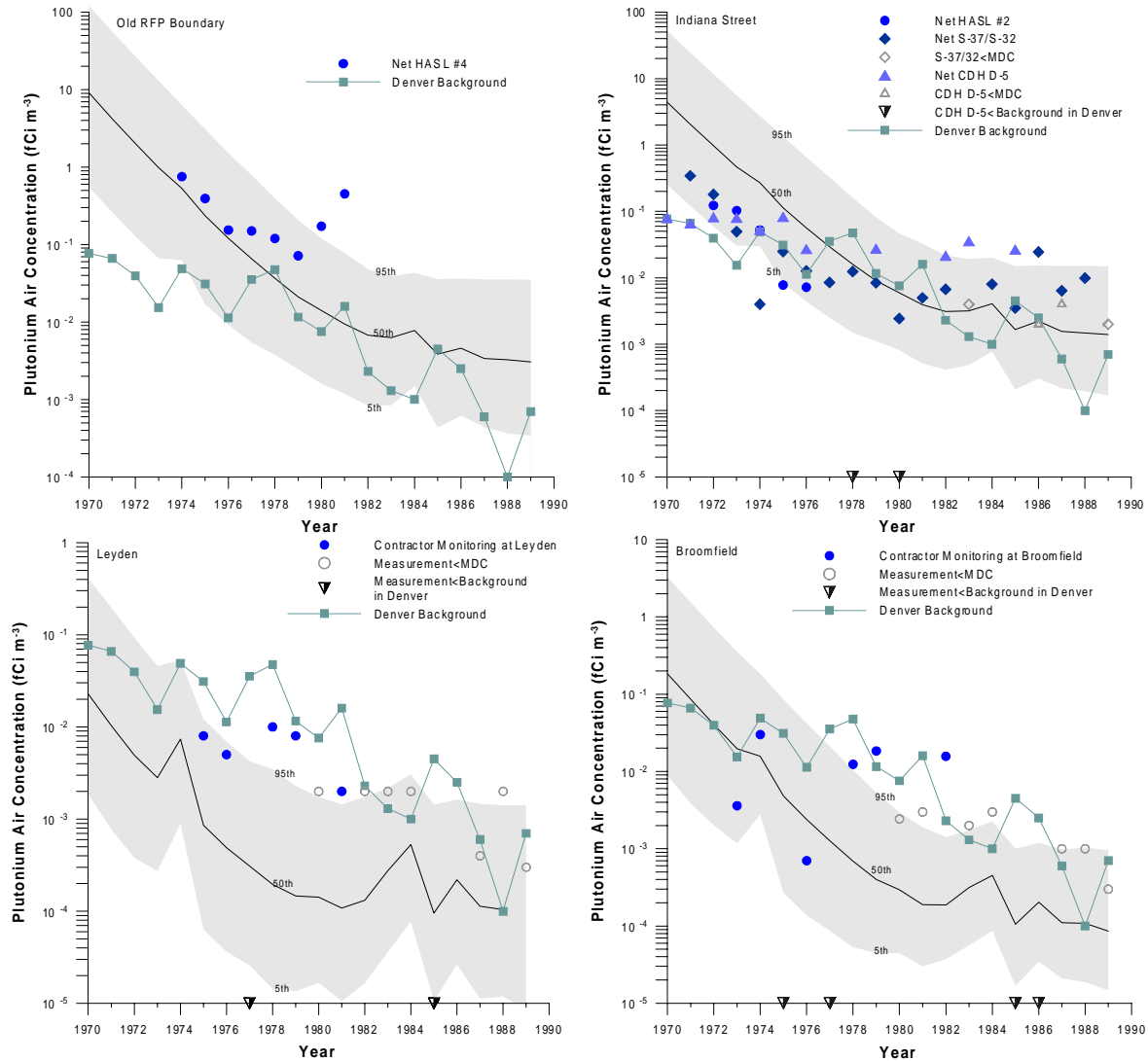
Comparisons of predicted and measured plutonium air concentrations are presented in [Figure 19](#). Annual average background concentrations from fallout sources in Denver ([Rope et al. 1999](#)) are also plotted for comparison, although background was subtracted from the measured values. Note that fallout background concentrations in the late 1980s are near that of the net measured concentrations. Measurements where the annual average concentration was less than the fallout concentration are noted in the caption to [Figure 19](#). In addition, those measurements that should be viewed as more uncertain, because they were less than the agency's reported MDC, are shown with open (not solid) symbols in [Figure 19](#).

Predicted concentrations during the 1970s are dominated by resuspension sources, with some perturbations during routine releases from buildings and stacks. The peaks observed in the predicted concentrations in 1974 and 1984 reflect routine releases from the Building 771 stack and Building 776 roof vents. The decrease in the predicted concentrations of plutonium in air from 1970 to 1980 is in response to the decrease in resuspension over time and to a lesser extent, the weathering of plutonium from the 0–3-cm soil layer.

In general, model predictions encompass the measured values until about 1976. After 1976, the model tends to underpredict air concentrations at all locations except Indiana Street. At the Indiana Street location, the model underpredicts some of the measurements during the 1981 to 1988 time frame. There is a noticeable increase in the measured concentration during the years 1976 to ~1982 at all stations except Indiana Street. The reason for this increase was not determined. During the 1976–1982 time frame, several of the annual average concentrations at Leyden and Broomfield were either less than Denver background or less than the agency's MDC. These measurements should be interpreted with caution because of the large uncertainty associated with them. After 1983, all annual average concentrations at the Leyden and Broomfield stations were less than the RFP contractor's MDC. RFP contractor and CDPHE samplers at Indiana Street during the 1982–1989 time frame had several measurements less than the MDC.

It is interesting to note that the measured concentrations at the old RFP boundary between the years 1974–1979 (HASL 4) fall off with about the same slope as the model-predicted values. This observation provides some validation of the time-dependent soil resuspension model used in this study. In 1980–1981, measured concentrations at HASL 4 increased substantially. One possible reason is that the HASL 4 air sampler may have been picking up large particles of suspended contamination from vehicular traffic. Inspection of the HASL 4 and HASL 1 (near 903 Area) samplers in 1981 showed that they were subject to gross contamination by dust from the dirt roads ([Feely et al. 1985](#)). The samplers were at ground level and only a meter or so away from the dirt roads. The wooden louvered shelters did not prevent dust raised by passing vehicles from settling on the samplers and on the exposed filters. It seemed likely that a large part of the aerosol being collected was resuspended coarse dust, not necessarily representative of the aerosol that would have been collected if the sampler had been sitting on a metal stand, as were most of the samplers in their Surface Air Sampling Program. Sampler HASL 2 at Indiana Street was not included in the [Feely et al. \(1985\)](#) discussion on this topic; therefore, the proximity to suspended contamination from an unpaved surface does not appear to have been a concern at the sampler location. The HASL 2 measurements fall in line with the lower percentiles of the predicted

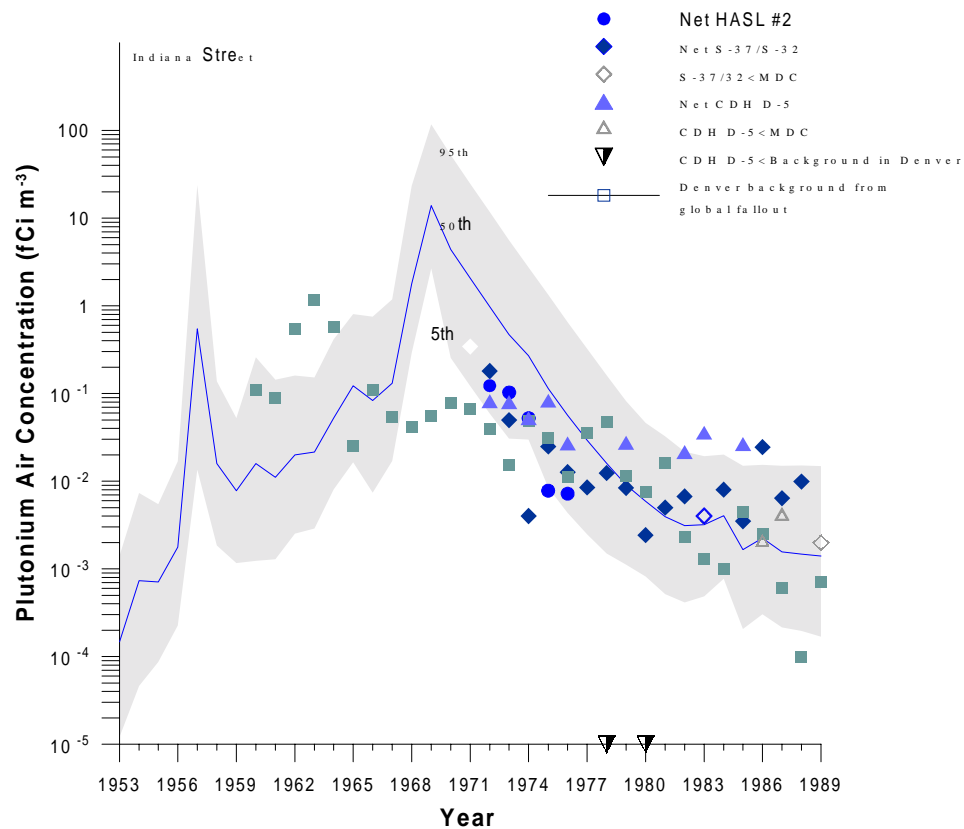
values and with measurements made by the RFP contractor and the CDPHE at Indiana Street (Figure 19).



**Figure 19.** Comparison of predicted annual average plutonium concentrations in ambient air with measurements at four locations in the model domain. Measured concentrations in Denver are also shown and are consistent with global fallout concentrations in other U.S. cities. This estimate of the fallout plutonium concentration was subtracted from the annual average measured value at the four locations so that net concentrations above background are plotted. Years where the measured concentration of plutonium in air was less than background are shown. Those years were 1978 and 1980 for CDH D-5; 1975, 1977, 1985, and 1986 for Broomfield; and 1977 and 1985 for Leyden. Open symbols in the plots indicate measurements that are quite uncertain because the measured annual average concentration was less than the minimum detectable concentration for that agency.



It is obvious from [Figure 19](#) that the model fails to account for all important processes that would influence air concentrations at a sampler. These processes may include enhanced resuspension from vehicle traffic, human activity, and high winds, as well as transport of plutonium from upwind resuspension sources. However, it is important to put these concentrations into perspective in terms of the overall magnitude of exposure from RFP and fallout sources during the operational period of the RFP (1953–1989). Figure 20 shows the predicted annual average air concentration at Indiana Street from 1953–1989, along with measured data taken there after 1970. Note that the peak annual average concentrations were about 1 to 2 orders of magnitude greater than concentrations after 1970, and during some years, background concentrations exceeded predicted concentrations from RFP sources.



**Figure 20.** Predicted annual average plutonium concentrations in ambient air from 1953 to 1989 at Indiana Street and plutonium-specific measurements taken from 1970 to 1989. Plutonium from global fallout and measured in Denver (shown separately) has been subtracted from the measured values. Estimated plutonium background in Denver from global fallout is also shown.

### Model Comparisons with Data Taken During the 1969 Fire

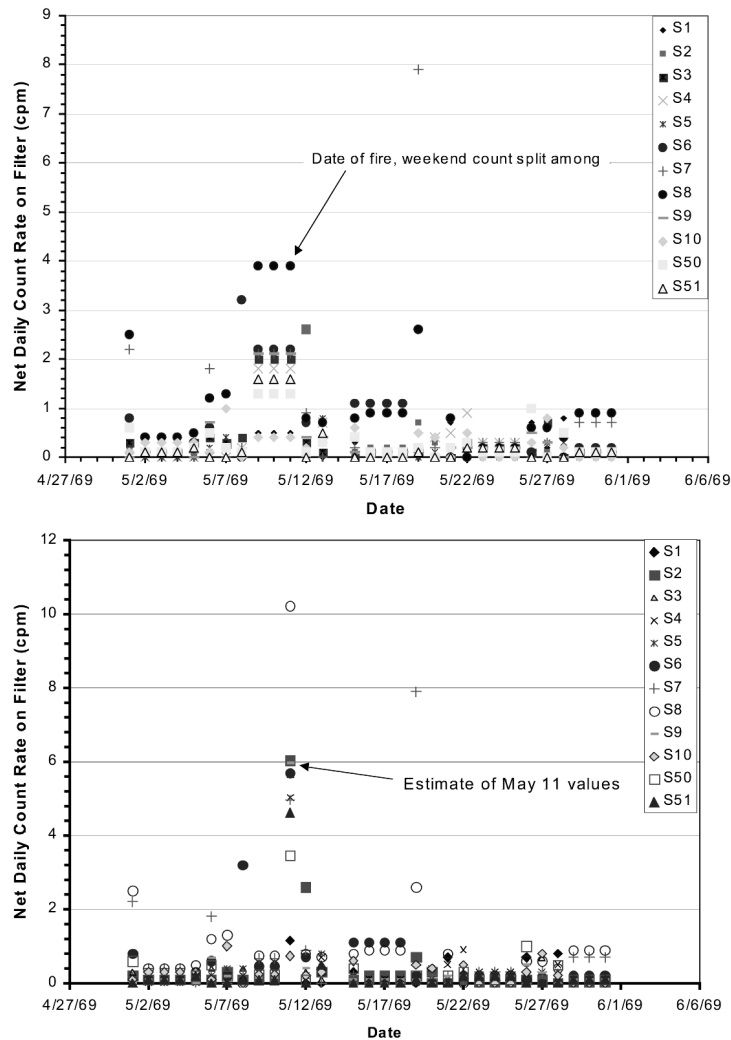
Predicted 24-hour average plutonium concentrations within the RFP industrial area were compared with *estimated* plutonium concentrations based on onsite sampler results of TLLα activity. The fire occurred on Sunday, May 11, 1969, in Building 776. The standard practice at that time for sampling over a weekend was to allow the sampler to run from 8:15 a.m. Friday to

8:15 a.m. the following Monday. The results were then reported for the 3-day sampling period. To arrive at the estimated count rate (in counts per minute) of TLL $\alpha$  activity for the day of the fire, we first calculated the average net daily count rate for May, excluding those data that were taken over the weekend the fire occurred (Table 17). The daily averages were then multiplied by 2 (to represent the 2 prefire days in the 3-day sampling period) and subtracted from the net count rate observed on the filter paper for the weekend that included May 9, 10, and 11. The new estimated counts for May 11 were then converted to activity concentrations in air using the conversion factor derived by [Rope et al.](#) (1999) of 0.038 cpm (fCi m<sup>-3</sup>)<sup>-1</sup>. All excess TLL $\alpha$  activity was assumed to be plutonium that originated from the fire.

The time trend in daily net count rate for the month of May 1969 is shown in [Figure 21](#). The upper graph in [Figure 21](#) shows the raw data. Note that for each 3-day weekend period, all three days have the same count rate. That is because, in our database of TLL $\alpha$  activity in onsite air, we divided the total count equally among the sampled days. Also note that the weekend of May 9, 10, and 11 shows substantially higher count rates than the other days. The lower graph shows the same data but the weekend of May 9, 10, and 11 has been segregated into estimates for the three individual days. Particle size of the effluent was assumed to be in the submicron range and typical of HEPA-filtered effluent. Sampler inlet collection efficiency for 1-micron particles was reported to be 100% ([Rope et al.](#) 1999), and we assumed the same efficiency for submicron particles in this exercise.

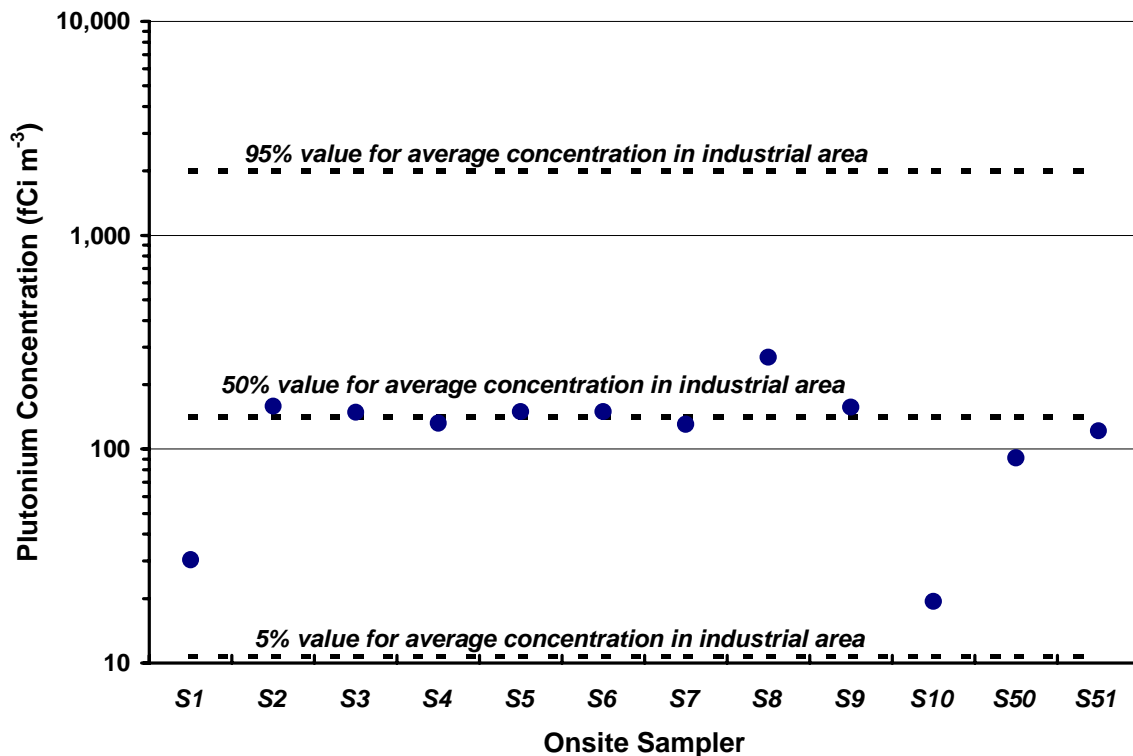
**Table 17. Measured Data at Onsite Samplers for the Weekend of May 9–11, 1969**

Sampler	Daily average count rate for May 1969 (cpm)	Corrected count rate for May 9, 10, 11, 1969	Estimated 24-hour average plutonium concentration in air for May 11, 1969
		(cpm)	(fCi m <sup>-3</sup> )
S1	0.173	1.5	30.36
S2	0.131	6.3	158.91
S3	0.173	6	148.79
S4	0.185	5.4	132.39
S5	0.154	6	149.80
S6	0.454	6.6	149.80
S7	0.665	6.3	130.77
S8	0.738	11.7	269.03
S9	0.162	6.3	157.29
S10	0.231	1.2	19.43
S50	0.219	3.9	91.09
S51	0.088	4.8	121.66



**Figure 21.** Net daily count rates for the month of May 1969. The upper graph shows the average daily count rate for the weekend of May 9, 10, and 11. In the lower graph, the count rate for May 11, 1969, has been segregated from May 9th and 10th.

We calculated the predicted 24-hour average plutonium concentration in air at the six model nodes nearest to the RFP industrial area boundaries. Three of the nodes were on the north, south, and southwest industrial area boundary. The remaining three nodes were located just outside the northwest, southeast, and northeast industrial area boundary (see [Figure 3](#)). No nodes were present in the interior of the industrial area. The average concentration at these six nodes was compared to the estimated measured concentrations from the onsite samplers ([Figure 22](#)). No corrections were made for background plutonium concentrations from fallout sources. This would have resulted in an insignificant correction, as fallout background in Denver in 1969 was  $0.07 \text{ fCi m}^{-3}$ , much less than the MDC for the onsite samplers. As shown in [Figure 22](#), median



**Figure 22.** Predicted and measured 24-hour average plutonium concentration in ambient air in the RFP industrial area for May 11, 1969.

(50%) predicted concentration values matched the sampler data reasonably well. In fact, all measurements were within the 5th to 95th percentile range.

The good agreement between the predicted and measured values is not interpreted as convincing evidence that the model has predicted concentrations offsite accurately. One problem with the comparison is that building wake was not included in the model. Building wakes will tend to dilute concentrations in some areas, while enhancing concentrations at others. Without a detailed evaluation of wake effects that takes into consideration all buildings in the industrial area, it is difficult to say whether the model underpredicted or overpredicted concentrations. Despite these shortcomings, the comparison does provide some evidence that the dispersion modeling and source term are at least reasonable estimates of what might have been released and transported during the fire.

### Model Comparisons with Lake and Reservoir Sediment Core Data

Lake sediment, like soil, is an environmental medium in which plutonium may accumulate over time. Unlike soil, contamination tends to accumulate in bottom sediments in discrete layers that build upon the previous layer. There is little or no vertical migration of radionuclides between undisturbed sediment layers. Thus, the temporal history of contaminant deposition can be preserved in the sediment.

An overview of sediment monitoring studies conducted near Rocky Flats is given in Chapter VII of [Rope et al.](#) (1999). Some routine monitoring of sediment for radionuclides was conducted by the RFP contractor in the early 1950s and 1970s. Generally, however, that routine monitoring involved few sampling sites, undocumented methods, and infrequent reporting. Therefore, these data are of limited use. Studies that were more comprehensive were performed from 1969–1992 by various agencies including the EPA, Colorado State University, Battelle Pacific Northwest Laboratories, Colorado School of Mines, City of Broomfield, U.S. Geological Survey, and Environmental Measurements Laboratory<sup>d</sup>. Plutonium-239/240, <sup>89,90</sup>Sr, <sup>137</sup>Cs, <sup>241</sup>Am, and naturally occurring radionuclides were analyzed during these studies.

Analysis of sediment cores has allowed for evaluating temporal trends of radioactivity in this medium because sediment layers can be age-dated. Unfortunately, the airborne component of this activity may be somewhat obscured by liquid effluent discharges. For example, in 1972, reconstruction of some of the holding ponds at Rocky Flats led to increases in measured activity in sediment in Walnut Creek and Great Western Reservoir. Later studies revealed that the activity in Great Western Reservoir could have two components: atmospheric fallout and waterborne releases from the RFP. The atmospheric component was linked by age-dating the sediments to the time when high releases from the 903 Area occurred. Similar studies on Standley Lake also indicated high plutonium concentrations that correspond to the time of highest 903 Area releases.

Liquid effluent from the RFP was discharged into Woman Creek and North and South Walnut Creeks ([Rope et al.](#) 1999). Woman Creek drains into Standley Lake, and North and South Walnut Creeks drain into Great Western Reservoir. Effluent was first discharged to holding ponds to allow radioactivity to settle out before draining, although in the early 1950s, low-level contaminated laundry waste went directly into North Walnut Creek. Ponds B-1–B-5 received decontamination wastewater, laundry wastewater, and sanitary wastes and discharged to South Walnut Creek. Ponds A-1–A-4 received contaminated laundry wastewater, cooling tower blowdown, and condensate and discharged to North Walnut Creek. These ponds were constructed in the 1950s and underwent major reconstruction during the early 1970s. The reconstruction is suspected to have caused the elevated levels of plutonium observed in liquid effluent monitoring during this time. Discharges to these ponds were routinely monitored from the early 1950s.

Ponds C-1 and C-2 received filter backwash effluent and cooling tower blowdown. We suspect the RFP contractor did not think these sources contained significant radioactivity because they were not routinely monitored. For this reason, the activity in Standley Lake is thought to be mainly from deposition from airborne plumes and surface erosion of contaminated soil. Beginning in 1970, isotope-specific monitoring began in all three creeks.

For model comparisons, we selected a data set consisting of two sediment cores collected from the center of Standley Lake and reported in [Hardy et al.](#) (1978). While the radioactivity in liquid effluent discharged to Woman Creek is suspected to be minor, surface erosion and subsequent fluvial transport of plutonium-contaminated soil may have also contributed to the plutonium observed the Standley Lake sediment cores. We did not include surface erosion and fluvial transport in the model. Therefore, it is likely that the model will underpredict concentrations in lake sediments.

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<sup>d</sup> Formerly the HASL.

Data from Great Western Reservoir were considered; however, interpretation of these data is complicated by liquid effluent discharges to North and South Walnut Creeks. While the timing of 903 Area releases could be identified from Great Western Reservoir core data, it was difficult to say how much of the plutonium was from liquid effluent or airborne sources. [Schoep and Whicker](#) (1995), however, estimated that waterborne releases were responsible for about 87% of the *total* plutonium in Great Western Reservoir. Therefore, model-predicted inventory in Great Western Reservoir was compared with the estimated plutonium inventory that was attributed to airborne sources.

### Model Comparisons with Standley Lake Core Data

The two sediment cores collected and reported in [Hardy et al.](#) (1978) were analyzed for both  $^{137}\text{Cs}$  and transuranics, including  $^{239/240}\text{Pu}$  and  $^{238}\text{Pu}$ . Sedimentation rates were determined by correlating the concentration at different depths in the sediment core with global fallout of  $^{137}\text{Cs}$  from weapons tests and  $^{238}\text{Pu}$  from the SNAP-9A satellite. A sedimentation rate of  $3.4 \text{ cm y}^{-1}$  was determined from these data. Global fallout plutonium was differentiated from Rocky Flats plutonium through mass isotopic ratios of  $^{239}\text{Pu}$  to  $^{240}\text{Pu}$  ([Table 18](#)). Concentrations of Rocky Flats plutonium in lake sediment were reported in terms of activity per square meter. These values were converted to activity concentration per unit mass of dry sediment by dividing by the dry sediment density measured in [Hardy et al.](#) (1978).

The model did not include lake sediments as a specific medium in the model domain, and the area covered by Standley Lake was treated like soil. Using the option in the model that allows for output of the deposition for a single year, model-predicted concentrations in the 0–3-cm soil layer for each year from 1964 to 1972 were obtained at the four nodes bounded by Standley Lake. Recall that for year in which deposition occurred, all deposited plutonium resided in the 0–3-cm layer. We first calculated the average concentration of the four nodes. We then converted the average concentrations of the four nodes to sediment concentrations by adjusting for differences between the soil and lake sediment density and averaging thickness as shown in Equation (20).

$$C_{sed} = C_{soil} \frac{\rho_{soil} T_{soil}}{\rho_{sed} T_{sed}} \quad (20)$$

where

- $C_{sed}$  = plutonium concentration in dry lake sediment ( $\text{Ci g}^{-1}$ )
- $C_{soil}$  = predicted plutonium concentration in the top 3-cm of soil at nodes bounded by Standley Lake for the year deposition occurred ( $SC$  in [Equation 8](#) [ $\text{Ci g}^{-1}$ ])
- $\rho_{soil}$  = average soil density in 0–3 cm soil layer ( $\text{g cm}^{-3}$ )
- $\rho_{sed}$  = dry lake sediment density ( $\text{g cm}^{-3}$ )
- $T_{soil}$  = averaging thickness of soil layer (3 cm)
- $T_{sed}$  = annual sediment accumulation (3.4 cm).

The dry lake sediment density was obtained from data in the 5th column of [Table 18](#) for each respective year of deposition. These values were reported in areal density ( $\text{g cm}^{-2}$ ) and were converted to a volume density ( $\text{g cm}^{-3}$ ) by dividing by the sediment core thickness (2 cm).

**Table 18. Plutonium in Standley Lake Sediment Measured by [Hardy et al. \(1978\)](#)**

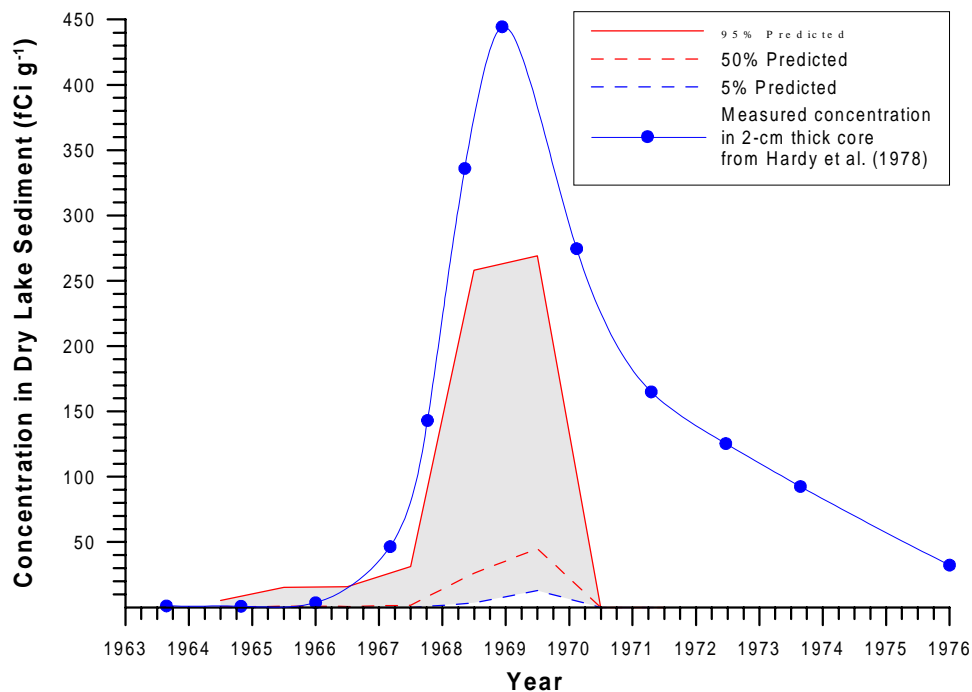
Depth increment (cm)	Total plutonium activity (nCi m <sup>-2</sup> )	Global plutonium fallout (nCi m <sup>-2</sup> )	Rocky Flats plutonium (nCi m <sup>-2</sup> )	Measured dry density (g cm <sup>-2</sup> ) <sup>a</sup>	Rocky Flats plutonium (fCi g <sup>-1</sup> )	Estimated year of deposit <sup>b</sup>
0–2	0.31	0.08	0.23	0.71	32.4	1976
8–10	0.58	0.08	0.50	0.54	92.6	1973.7
12–14	0.86	0.12	0.74	0.59	125	1972.5
16–18	1.22	0.23	0.99	0.6	165	1971.3
20–22	1.93	0.20	1.73	0.63	275	1970.1
24–26	3.57	0.37	3.2	0.72	444	1968.9
26–28	2.87	0.35	2.52	0.75	336	1968.4
28–30	1.46	0.43	1.03	0.72	143	1967.8
30–32	0.66	0.32	0.34	0.73	46.6	1967.2
34–36	0.22	0.19	0.03	0.83	3.61	1966
38–40	0.59	0.58	0.01	1.1	0.909	1964.8
42–44	0.66	0.65	0.01	0.84	1.19	1963.6

<sup>a</sup> Areal density per 2-cm thick core segment.<sup>b</sup> Based the dividing the top of the core segment by the average sedimentation rate (3.4 cm y<sup>-1</sup>) and subtracting that value from the 1976 value.

Predicted and measured concentrations of plutonium in Standley Lake sediment cores ([Figure 23](#)) showed that the model underpredicted concentrations in lake sediments from 1967 to 1976. The model underestimated the peak concentration in 1969 by about a factor of 2 at the 95% level and almost a factor of 5 at the 50% level. This result is puzzling, because the model predicted the soil concentration at the [Webb](#) (1996) sampling site, FX3 (see [Table 15](#) and [Figure 16](#), 120° transect), which is near western shore of Standley Lake, was within the range of the measured value. However, model-predicted soil concentrations at the Poet and Martell sampling sites near Standley Lake (site V and N, see [Table 16](#) and [Figure 17](#)) were underpredicted. Predicted concentrations at these sampling sites were slightly greater than the 95th percentile predicted value. Sampling site V is near Webb's sampling site FX5, and soil concentrations showed a 7-fold decrease between 1970 and 1994 (0.24 pCi g<sup>-1</sup> in 1970 compared to 0.034 pCi g<sup>-1</sup> in 1994), the years the Poet and Martell and Webb sampling were performed, respectively. We note that [Poet and Martell](#) (1972) sampled the 0–1-cm depth while [Webb](#) (1996) sampled from 0–3 cm. However, accounting for the sampling difference and downward migration of plutonium in the soil column still does not account for the 7-fold decrease in concentration. Based on this information, we suspect the model underpredicted deposition in the Standley Lake area but not to the extent as indicated by the sediment core data.

Two processes not included in the model are thought to have caused the apparent underestimation of plutonium concentrations in Standley Lake sediments: (1) erosion and fluvial transport of contaminated soil and (2) airborne transport of resuspended contaminated soil. Surface erosion includes processes such as sheet wash, in which water flows freely over the land surface and transports fine soil particles as it moves. These waters may eventually drain into





**Figure 23.** Predicted and measured concentration of Rocky Flats plutonium in annual accumulations of dry lake sediment in Standley Lake. Measured data were obtained from [Hardy et al. \(1978\)](#); global fallout plutonium was subtracted. Each year is equivalent to 3.4 cm of sediment.

Woman Creek or directly into Standley Lake to be incorporated into the sediment bed. Differences in the surface soil plutonium concentration observed by [Poet and Martell \(1972\)](#) and [Webb \(1996\)](#) could have been caused by surface erosion. Airborne transport of resuspended contaminated soil was not included in the model; instead, resuspended contaminated soil was assumed to deposit near its source. Soil concentrations were not affected by this process. For resuspension to affect lake sediment concentrations, resuspended contaminated soil would have to be transported from its point of origin and deposit in the lake.

Surface erosion and transport of resuspended contaminated soil are also believed to be partially responsible for the shape of the measured concentration curve from 1970 to 1976. Major airborne releases of suspended plutonium from the 903 Area ceased after it was paved in late 1969. The effect of this is observed in the predicted concentration curve that drops off steeply after 1969. However, the measured concentration curve drops off more gradually during this period, indicating a slowly declining source of plutonium not included in the model. This slow drop off in concentration is suspected to be from surface erosion and deposition of resuspended contaminated soil. Over time, surface soil contamination moved into deeper soil layers, making it less susceptible to surface erosion and resuspension.

One other observation of interest is worth pointing out in [Figure 23](#). The predicted concentration in sediments for 1968 (95th percentile value) are almost the same as the predicted concentration for 1969 (95th percentile value). However, predicted releases from the 903 Area were substantially higher in 1969. To understand this difference, we plotted surface deposition

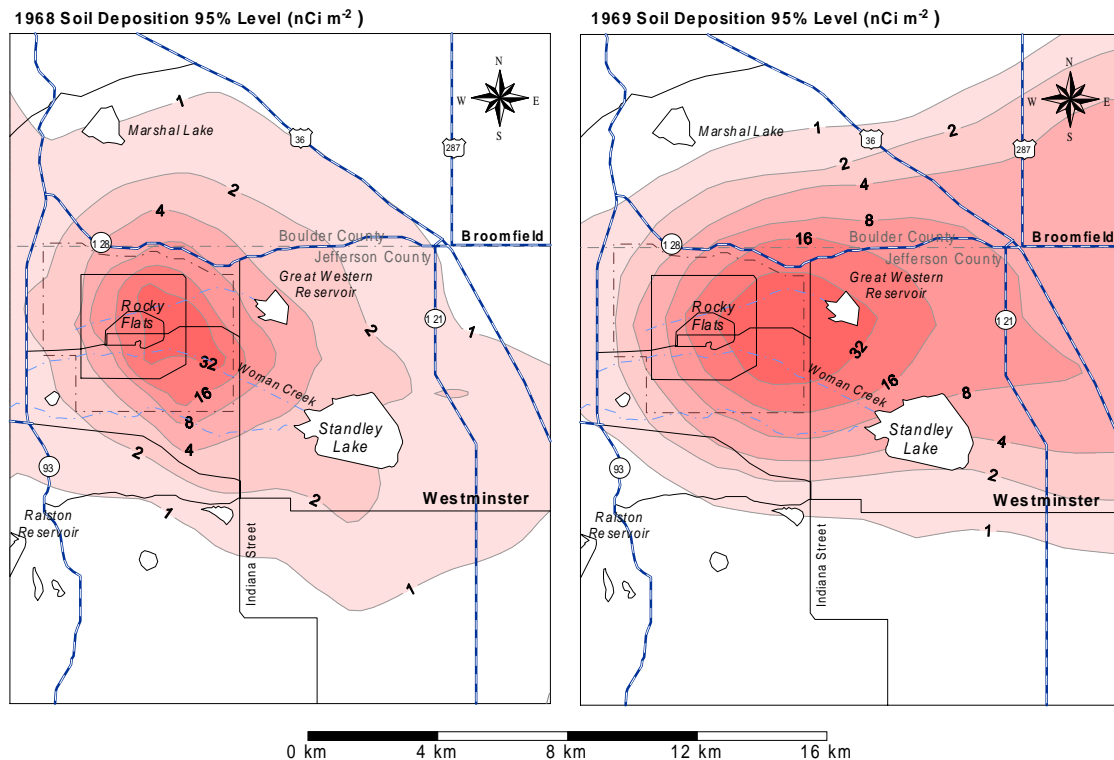


patterns for 1968 and 1969 (Figure 24). For 1968 (left plot), most of the deposition was from 903 Area baseline releases because only one discrete event was included in that year. Contrast that with 1969 (right plot), where most of the releases were from discrete events and surface deposition was substantially greater. Deposition patterns trend east-southeast in 1968 but trend east to northeast in 1969. The east-northeast trend is due to the wind direction estimated for each discrete event day. Note that surface deposition for 1968 is about the same for 1969 in the Standley Lake area.

### Model Comparisons with Inventory Estimates in Great Western Reservoir

Researchers from Battelle Pacific Northwest Laboratories ([Thomas and Robertson 1981](#)) and Colorado State University ([Schoep and Whicker 1995](#)) estimated plutonium inventories in Great Western Reservoir. These and other sediment studies are discussed in Chapter VII of [Rope et al. \(1999\)](#) (see [Table 19](#)).

[Thomas and Robertson \(1981\)](#) made their inventory estimates by first dividing the reservoir into three zones of plutonium activity and sediment thickness. Plutonium concentrations with depth were estimated for each zone based on eight core samples. Integrating across the three zones and summing yielded a total plutonium inventory estimate of 244 mCi. No correction for global fallout was apparently made, but this correction would have made little difference to the overall estimate.



**Figure 24.** Predicted plutonium deposition patterns at the 95% level for releases in 1968 (left plot) and 1969 (right plot). All sources active at the time were included in the simulation. Deposition is illustrated for the given year only.

**Table 19. Predicted and Estimated Plutonium Inventories in  
Great Western Reservoir Sediments**

Source of estimate	Total inventory in reservoir (Ci)	Inventory attributed to airborne sources (Ci)
<a href="#">Thomas and Robertson</a> (1981)	244	32
<a href="#">Schoep and Whicker</a> (1995), Method 1	54	7
<a href="#">Schoep and Whicker</a> (1995), Method 2	62	8
<a href="#">Schoep and Whicker</a> (1995), Method 3	92	12
Model predicted 95%	---	53
Model predicted 50%	---	12
Model predicted 5%	---	4.6

[Schoep and Whicker](#) (1995) collected 48 sediment cores and estimated plutonium inventory using three different methods. The first method took the median activity per unit area in the reservoir ( $0.08 \mu\text{Ci m}^{-2}$ ) and multiplied it by the area of the reservoir ( $0.67 \text{ km}^2$ ), which yielded a plutonium inventory of 54 mCi. The second method divided the reservoir into 4-m contours and multiplied the median plutonium activity per unit area for the depth by the area enclosed within the contour. These values were then summed to yield a plutonium inventory of 62 mCi. The third method tried to duplicate the method of [Thomas and Robertson](#) (1981). Using this method they estimated a plutonium inventory of 92 mCi compared to the 244 mCi calculated by [Thomas and Robertson](#) (1981). [Schoep and Whicker](#) (1995) also estimated the fraction of plutonium activity in the reservoir sediments that was attributed to aquatic pathways. Using ratios of plutonium to  $^{137}\text{Cs}$  in soil and sediment, they estimated about 87% of the plutonium in the reservoir was attributed to aquatic pathways. We used this value in deriving the plutonium inventory in the reservoir that was attributed to airborne releases.

Predicted plutonium inventory was estimated from the predicted soil concentration data for the single node that lies within Great Western Reservoir (see [Figure 3](#)). The predicted soil concentration in the 0–3-cm layer was first divided by the fraction of the total plutonium deposited that remained in that layer for the year 1989 (0.53). This fraction (0.53) assumed that most of the contamination was deposited in 1969. The corrected plutonium concentration was then multiplied by the 0–3-cm soil density ( $0.829 \text{ g cm}^{-3}$ ) and the layer thickness (3 cm) to yield the areal concentration of plutonium in the reservoir. We then multiplied the areal concentration by the area of the reservoir ( $0.67 \text{ km}^2$ ) to obtain the inventory.

Results of the comparison ([Table 19](#)) showed that inventory estimates based on core data were within the 5th and 95th percentile model predictions. Estimates made by the first two methods employed by [Schoep and Whicker](#) (1995) were close to the predicted inventory at the 5th percentile level, suggesting a tendency of the model to overpredict concentrations in this area. Using the third method employed by Schoep and Whicker, the estimated inventory was close to the median (50%) predicted inventory. The inventory estimate made by [Thomas and Robertson](#) (1981) is less than the 95th percentile of the predicted inventory but greater than the median predicted inventory.

### Model Comparisons with Vegetation Monitoring

Vegetation is the only medium for which a significant number of plutonium-specific measurements were made before 1970. This medium may, therefore, be important for validating source terms and environmental transport modeling.

Vegetation monitoring studies are summarized and discussed in Chapter V of [Rope et al. \(1999\)](#). Monitoring of vegetation began before the site was operating as part of a preoperational background study. The background gross alpha activity in vegetation was estimated to be  $86 \pm 73$  pCi kg<sup>-1</sup> (one standard deviation). Initial monitoring began in 1952, but it ended in 1953 because of technical problems. Vegetation monitoring resumed after the 1957 fire and continued into 1958. The RFP contractor's site survey monthly reports, in which these data are reported, do not specify the types of plants that were collected or the portions of plants that were analyzed. It appears that the collection and analytical techniques were the same as those used in 1952 and 1953 (Hammond [1957](#), [1958](#)).

Routine offsite vegetation monitoring from 1963 to 1970 was performed by the RFP site contractor, Dow Chemical Company, and reported in the Environmental Survey reports. Measurements consisted of gross alpha analysis of samples collected at 55 to 65 locations. Special sampling was conducted after the fire on May 11, 1969. Along with routine samples, 22 samples were collected from 11 offsite locations and were analyzed for both gross alpha and plutonium.

After 1970, the routine vegetation sampling data were reported in the Annual Environmental Monitoring Reports published by Dow. Samples were collected semiannually in 1971 and 1972 and analyzed specifically for plutonium. Routine vegetation sampling was not done from 1973 to 1978. Beginning in 1979, a new program was implemented. This was a more detailed investigation that sampled three categories of vegetation (forb, annual grass, and perennial grass) and analyzed for specific plutonium isotopes. Vegetation samples were not collected after 1983.

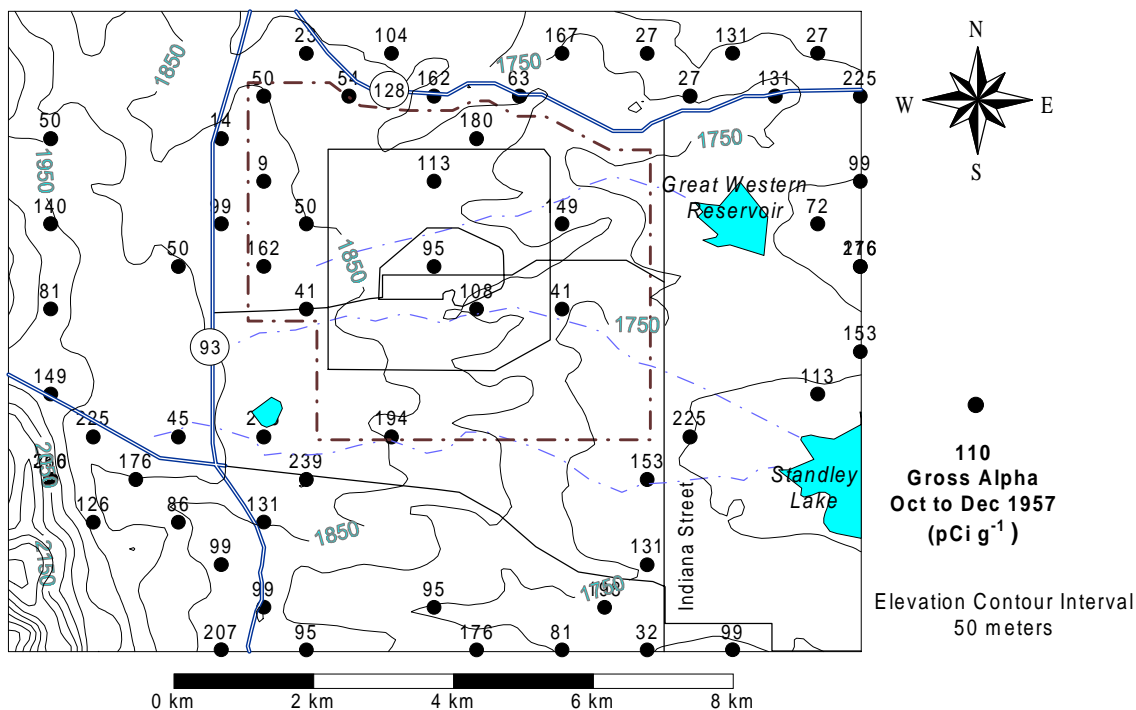
Vegetation is an important medium because many plutonium-specific measurements were made, and sampling was performed following significant release events such as the 1957 fire. However, comparisons of predicted-to-observed concentrations require additional modeling because vegetation was not a medium included in the model. Furthermore, the concentration on vegetation is highly variable, both temporally and spatially, and can be influenced by many different processes that include the vegetation interception fraction, surface weathering, resuspension, and rainsplash. These processes require detail about the type and part of vegetation sampled, the soil concentration beneath the plant, and soil conditions at the time of measurement. While the data taken from 1979–1983 appear to contain some of that detail, earlier measurements did not, making model comparisons difficult.

In general, we found more direct measurements of plutonium activity (such as in the air or soil) in the environment more useful for model comparison. Vegetation monitoring may be important for establishing general contamination trends, but predicting concentrations on vegetation is another matter, and it requires additional modeling beyond the exposure and risk needs for this project. For these reasons, we limited model comparisons to only those situations where no other environmental data exist. This restriction limited comparisons with vegetation data to the pre-1963 data.

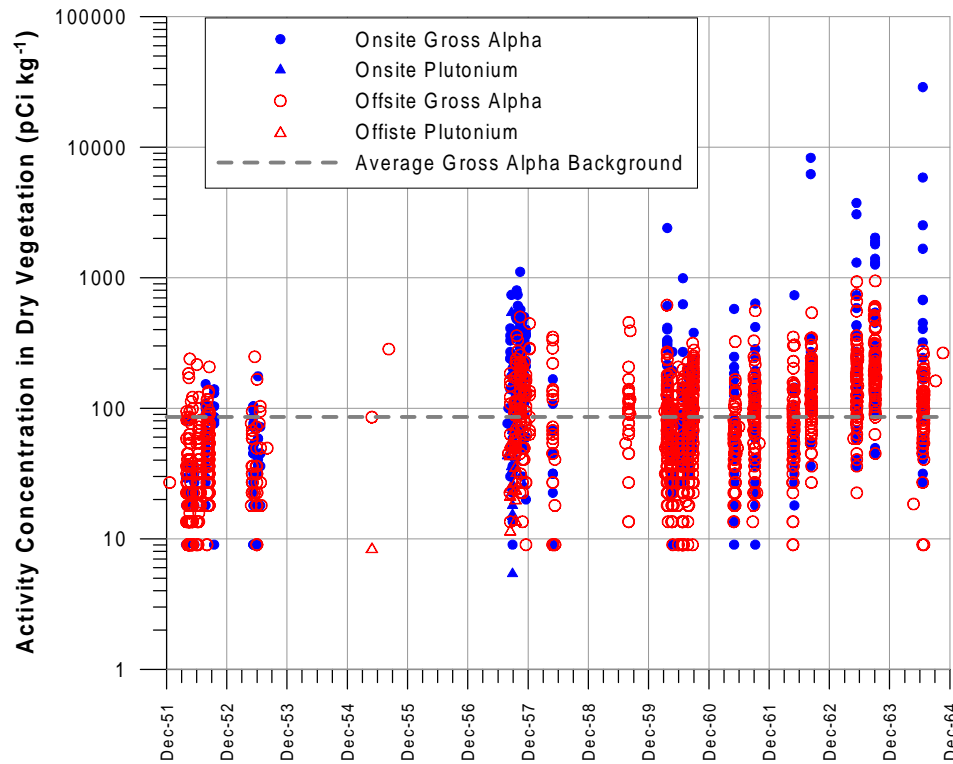
## Vegetation Sampling in Response to the 1957 Fire and Concentration Time Trends

In response to the 1957 fire, samples were collected at all locations established in the 1952–1953 vegetation monitoring program (Figure 25). The samples collected in conjunction with the fire included 222 vegetation samples in 1957 and 132 vegetation samples in 1958. Over 80 of the 1957 samples were analyzed specifically for plutonium. The data shown in Figure 25 represent only the gross alpha measurements made during one of the sampling surveys performed in 1957 following the fire. The Environmental Survey Reports present the average and maximum values for all samples collected within a given distance interval from the plant.

To understand the temporal trend of these data, gross alpha and plutonium measurements from 1952 to 1963 were plotted for samples taken onsite and offsite (Figure 26). Note that many of the samples fall below the average background value determined from the preoperational survey. There appears to be a general increase in the concentration over time, especially for samples taken onsite, but concentrations on any particular day sampled were found to vary by at least an order of magnitude across the sample study area. Concentrations in vegetation samples taken onsite may have been influenced more by resuspension from local contamination than from deposition from airborne plumes. Also note we do not see a substantial jump in the concentration for samples taken after the 1957 fire. However, concentrations are somewhat higher than background for this time. Few samples were taken between September 1953 and September 1955.



**Figure 25.** Gross alpha activity in vegetation measured after the September 11, 1957, fire in Building 771. Sampling was performed from September 13 to December 17, 1957. Measurements are not corrected for background activity levels or activity deposited before the fire.



**Figure 26.** Gross alpha activity concentration in vegetation onsite and offsite from 1952 to 1963 as measured by the Rocky Flats Plant site contractor. Each data point represents an individual measurement.

For model comparisons, we took the monthly average of offsite measurements in the sampling domain because model resolution was not refined enough to predict concentrations at a specific point and time. That is, we took all samples regardless of location collected during a given month and computed an average value. This average value was then compared with the corresponding model-predicted values. We omitted the onsite data from this comparison because (a) concentrations may have been more influenced by resuspension of local contamination derived from liquid or other effluent sources, (b) building wake effects may have resulted in enhanced local deposition (this process was not included in the model), and (c) concentrations offsite are of greater interest because this is where public exposure would have occurred.

### Model Predicted Concentrations on Vegetation

A simple model was constructed to predict the concentration on vegetation as a function of time. The conceptual model considers a time-varying deposition rate. A fraction of the activity deposited goes to the plant surface, and the remainder goes to the ground. Activity on the plant is weathered to the ground surface and activity on the ground surface is transferred back to the plant surface via resuspension and rainsplash. Root uptake was not considered a viable pathway because concentration ratios for plutonium are very small. The mass balance equation for plutonium on the plant surface per square meter of soil is given by

$$\frac{dQ_p}{dt} = FV R(t) - \lambda_w Q_p + (k_r + k_{rs}) Q_s \quad (21)$$

where

- $Q_p$  = plutonium on vegetation ( $\text{Ci m}^{-2}$ )  
 $FV$  = fraction of deposition that is intercepted and initially retained on vegetation  
 $R(t)$  = spatially-average surface deposition rate as a function of time ( $\text{Ci m}^{-2} \text{d}^{-1}$ )  
 $\lambda_w$  = weathering rate constant ( $\text{d}^{-1}$ )  
 $k_r$  = resuspension rate constant ( $\text{d}^{-1}$ )  
 $k_{rs}$  = rainsplash rate constant ( $\text{d}^{-1}$ )  
 $Q_s$  = plutonium on surface soil ( $\text{Ci m}^{-2}$ ).

Plutonium on the soil surface is given by

$$\frac{dQ_s}{dt} = (1 - FV) R(t) + \lambda_w Q_p - (k_r + k_{rs}) Q_s \quad (22)$$

The term  $R(t)$  represents plutonium deposition from RFP sources. The mean deposition of global fallout of plutonium for each respective year as reported in Appendix H of [Rope et al.](#) (1999) was also added to the term,  $R(t)$ , to account for background plutonium levels. Areal concentration on vegetation was converted to activity concentration in dry biomass by dividing the areal concentration by the standing dry biomass ( $\text{kg m}^{-2}$ ). The term,  $FV$ , is given by

$$FV = 1 - e^{-\alpha B} \quad (23)$$

where

- $\alpha$  = foliar interception constant ( $\text{m}^2 \text{kg}^{-1}$ )  
 $B$  = standing dry biomass ( $\text{kg m}^{-2}$ ).

We did not consider vegetation model parameters stochastically (Table 20), and we acknowledge that this will lead to an underestimation of uncertainty in the model output. Uncertainty was considered only in the average surface deposition rate over the sampling domain,  $R(t)$ . Parameter values for the vegetation model were derived from the dynamic food chain models, PATHWAY ([Whicker and Kirchner](#) 1987) and COMIDA ([Abbott and Rood](#) 1994).

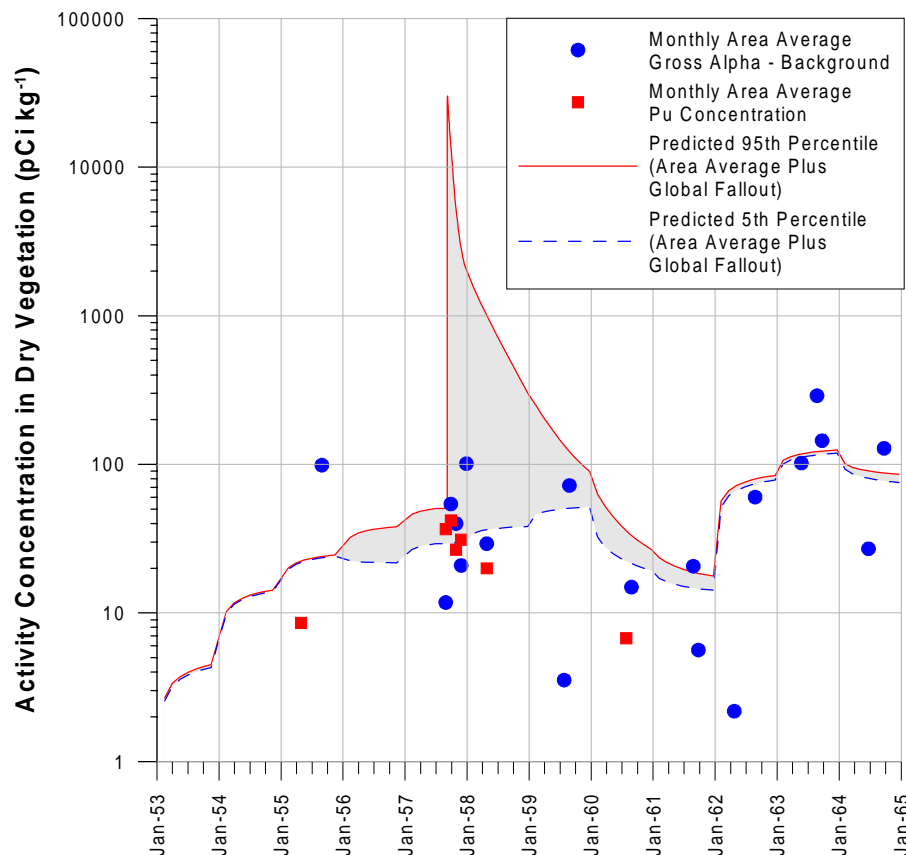
**Table 20. Vegetation Model Parameters**

Parameter	Value	Reference
Weathering rate constant	0.0495 $\text{d}^{-1}$	<a href="#">Whicker and Kirchner</a> (1987)
Resuspension rate constant	0.0017 $\text{d}^{-1}$	<a href="#">Whicker and Kirchner</a> (1987)
Rainsplash rate constant	0.00086 $\text{d}^{-1}$	<a href="#">Whicker and Kirchner</a> (1987)
Foliar interception constant <sup>a</sup>	3 $\text{m}^2 \text{kg}^{-1}$	<a href="#">Abbott and Rood</a> (1994)
Dry biomass <sup>b</sup>	0.1 $\text{kg m}^{-2}$	<a href="#">Abbott and Rood</a> (1994)

<sup>a</sup> Appropriate for particles <2  $\mu\text{m}$ .  
<sup>b</sup> The dry biomass of pasture grass was used.

Equations (21) and (22) were solved using a Runge Kutta numerical solver adapted from Press et al. (1992). Concentrations were output for the 5th and 95th percentile values and are shown in Figure 27 along with the monthly averaged measured values averaged over the sampling domain for offsite locations. The mean gross alpha background of  $86 \text{ pCi kg}^{-1}$  reported in Rope et al. (1999) was subtracted from each of the monthly average gross alpha counts to yield the gross alpha activity attributed to Rocky Flats sources. It is acknowledged that background is not a fixed value, and this subtraction results in some apparently positive measurements that are actually within the statistical range of background. Measured gross alpha concentrations less than the mean background are not shown in Figure 27.

Predicted concentrations before December 1955 were dominated by plutonium fallout, which accounts for the compression of the 5th and 95th percentile lines. After December 1955, predicted routine emissions exceeded global fallout. The most prominent trend observed in the measured concentrations was the drop in activity concentration (both gross alpha and plutonium) from the 1957 fire (September 1957) to December 1961. After that, measured concentrations generally began increasing in response to increased global fallout in 1962–1964. Predicted



**Figure 27.** Predicted and measured activity concentrations in vegetation from 1953 to 1965. Predicted concentrations represent  $^{239/240}\text{Pu}$  activity on vegetation averaged across the vegetation study area and include both RFP and global fallout sources. Measured gross alpha data are corrected for alpha activity from naturally occurring alpha emitters.



concentrations at the 5th and 95th percentile were again compressed from January 1962 to December 1964, but they responded to the increased global fallout. Predicted concentrations following the 1957 fire appeared to be biased high. The 95th percentile concentration for September 1957 was about a factor of 100 higher than the mean measured concentration of plutonium in vegetation.

It is difficult to interpret these data in light of the fact that activity concentrations on vegetation are highly variable and depend on localized conditions that were not included in the model. The general trends observed in the data set seem to support the weathering of activity from the plant surface following the 1957 fire, and they suggest either the 1957 fire source term was overestimated or dispersion modeling overpredicted concentrations in the vegetation study area. It is unlikely that deposition from routine releases was underestimated before 1957 based on the measurements. Measurements that exceed predicted concentrations in 1955 represent single measurements because only several samples were taken in vegetation during this time.

### **Summary of Model Validation**

Model validation exercises were performed using environmental data in four media: ambient air, soil, lake sediment, and vegetation. We used these comparisons to evaluate model accuracy and bias. In general, we found the comparisons supported the source terms and dispersion estimates used in the model. Even so, a number of model inadequacies were identified for which improvements can be made.

Comparisons with annual average ambient air measurements were hampered by lack of quality data before 1970. Consequently, comparisons were performed for post-1970 data only. Comparisons in this medium tested the resuspension portion of the model because releases after 1970 were dominated, for the most part, by resuspension of contaminated soil and not routine releases from the plant operations. Predicted concentrations onsite and at the new RFP boundary were generally within the range of measured values, with the exception of several years. The model underpredicted concentrations at community locations in the late 1970s and early 1980s. However, many of these measurements were below the agency's MDC and some measurements were less than background in Denver. Negative model bias (model underprediction) during the 1970s and 1980s will not substantially underestimate inhalation exposure for persons who lived in the model domain before this time because exposures were considerably less for those two decades than in the 1950s and 1960s.

Model-predicted soil concentrations 2 to 10 km east of the 903 Area were generally within the range of measured values. At distances <2 km from the 903 Area, the model underpredicted concentrations, while concentrations at distances >10 km were overpredicted. Model bias was most clearly seen in the 90° and 120° sampling transects used by [Webb \(1996\)](#). Much of the model bias was attributed to the particle size distribution of 903 Area releases. Modeled releases from the 903 Area did not include contaminated soil particles >30 µm. These particles are not an inhalation health risk because they are not respirable; however, they do contribute to offsite contamination. Overall, the distribution assigned to particles <30 µm appeared to have been biased toward the finer particles. That is, a greater fraction of the activity was assumed to be associated with smaller particles than the soil concentration measurements suggest. Larger particles would deposit closer to the source, resulting in higher predicted soil concentrations close to the source (the 903 Area) and lower predicted soil concentrations at greater distances.

Calibrating the particle size to deposition patterns will likely result in lower inhalation exposure because a greater fraction of the activity would be associated with larger, nonrespirable particles.

Model comparisons with estimates of plutonium soil inventory suggest that while the predicted spatial distribution of plutonium activity in soil did not correlate exactly with the measured data, the total amount of plutonium deposited in the model domain was reasonable and comparable with estimates made by other researchers.

Predicted concentrations in Standley Lake sediments were underestimated. This result was attributed to underestimation of deposition in the vicinity of Standley Lake and soil erosion and fluvial process that were not included in the model. Predicted plutonium inventory in Great Western Reservoir from airborne plumes matched values estimated by [Schoep and Whicker](#) (1995) and [Thomas and Robertson](#) (1981) reasonably well.

Predicted concentration in vegetation generally matched the temporal trends observed in vegetation measurements taken between 1953 and 1964. The model used to predict concentrations in vegetation appeared to overpredict concentrations from the 1957 fire. However, a lack of specific data about vegetation sampling methods and uncertainty in the vegetation model itself made model comparisons in this medium somewhat tenuous.

## INCREMENTAL LIFETIME CANCER INCIDENCE RISK ESTIMATES

This section presents isopleth maps showing the spatial distribution of the total (lung, liver, bone surface and bone marrow)<sup>e</sup> incremental lifetime cancer incidence risk for plutonium inhalation for each of the five receptor scenarios (Figures [28](#) through [32](#)). Individual organ risks were highest for the lung, followed by the liver, bone surface, and bone marrow. Isopleth maps were developed for the 5th and 95th percentile values of the output distributions. As noted in the discussion on model uncertainty earlier in this report, these percentiles represent the 95% confidence interval around the 5th and 95th percentile values.

The laborer ([Figure 28](#)) had the highest risk of all scenarios. This is not surprising because the laborer was assumed to have lived in the model domain for the entire period the RFP operated and had the highest breathing rate of any of the hypothetical receptors. Maximum incremental lifetime cancer incidence risks were in the  $10^{-4}$  range or a 1 in 10,000 chance of developing cancer during a lifetime. The area encompassed by the  $1 \times 10^{-4}$  risk level at the 95th percentile level extended south of the plant to the intersection of Colorado 58 and Interstate 70. At the 5th percentile level, the maximum cancer risk was in the  $10^{-7}$  range or a 1 in 10 million chance of developing cancer over his lifetime. A similar pattern is seen in the risk isopleths for the homemaker ([Figure 29](#)) and child scenarios ([Figure 30](#)). However, cancer risks are smaller because the breathing rates, and therefore, plutonium intake for these receptors were lower and exposure time for the child was shorter.

The office worker ([Figure 31](#)) was exposed from 1965 to 1989, and risk isopleths show a pattern different from that of the laborer, homemaker, and child scenarios. This difference arises because the office worker was assumed not to be present in the model domain during the 1957 fire. Maximum risks at the 95th percentile level and outside the RFP boundary at Indiana Street were around  $5 \times 10^{-5}$  or a 5 in 100,000 chance of developing cancer over their lifetime. The  $5 \times$

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<sup>e</sup> The lung, liver, bone surface, and bone marrow account for ~97% of the total risk from plutonium inhalation ([Grogan et al.](#) 1999)

$10^{-5}$  cancer risk encompasses a relatively small area east of Indiana Street. Cancer risks for this scenario were dominated by releases from the 903 Area. The student scenario ([Figure 32](#)), with an exposure period from 1964 to 1974, exhibits a similar pattern of risk to that of the office worker. However, risks are somewhat higher. The higher risks are attributed to an additional year of exposure to 903 Area releases (1964) and a higher breathing rate for the student compared to that of the office worker.

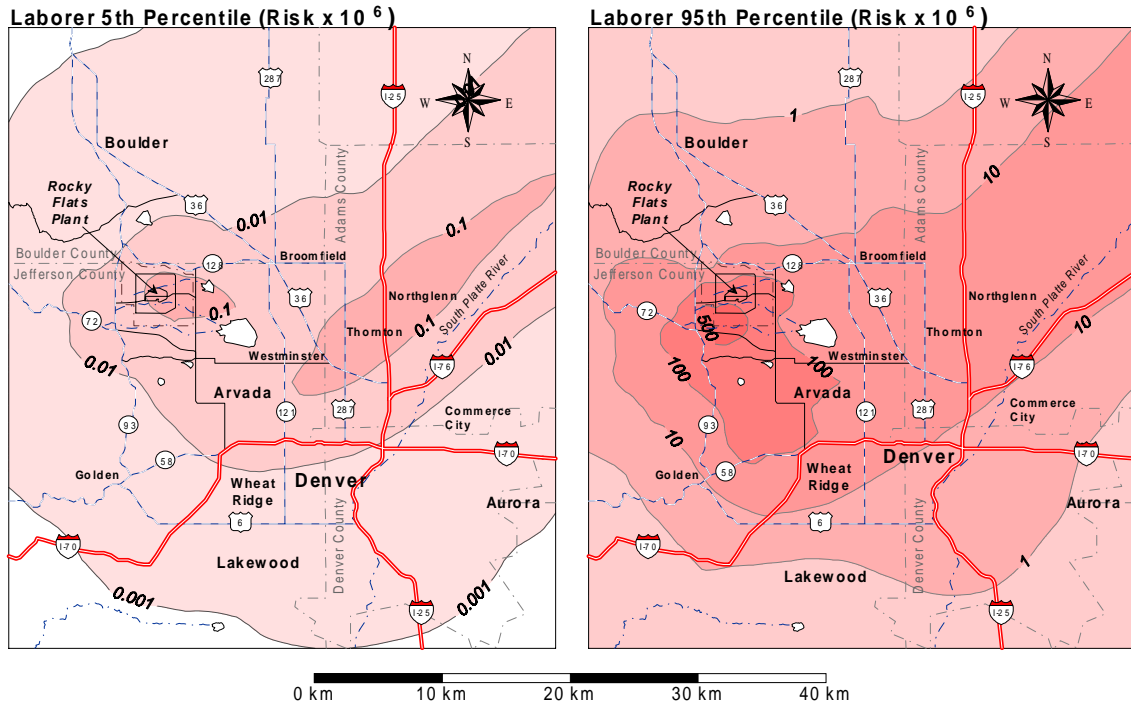
To understand the importance of receptor location and time of exposure, we extracted cancer risk estimates by decade of exposure at selected locations in the model domain ([Figure 33](#)) from model output for the laborer scenario ([Figure 34](#)). The relative importance of each decade of exposure depended on the location of the receptor and the percentile level chosen. Maximum risks at the 95th percentile were dominated by exposure during the years 1953–1959, provided the receptor was located in the plume path of the 1957 fire. Receptor locations outside the 1957 fire plume path were Denver, Boulder, Broomfield and Superior. Cancer risk estimates at these and other locations outside the 1957 plume path were highest during the 1960s in response to releases from the 903 Area. In most cases, the 50th percentile and 5th percentile followed this trend. However, at the Coal Creek location, maximum risk at the 50th percentile level occurred during the 1960s; at the 95th percentile level, maximum risk was incurred during the 1950s. Note the large uncertainty in the estimated risk from exposure incurred during the 1950s for the laborer located at Coal Creek. This uncertainty reflects the uncertainty in the 1957 fire plume path. While there is a low probability the plume traveled in this direction, the consequences of this happening are high.

Cancer risk estimates for the laborer scenario at the receptor locations illustrated in [Figure 33](#) are tabulated for the 5th, 50th, and 95th percentiles in [Table 21](#). Using the laborer located at Leyden as an example, the cancer risk estimates can be interpreted as follows:

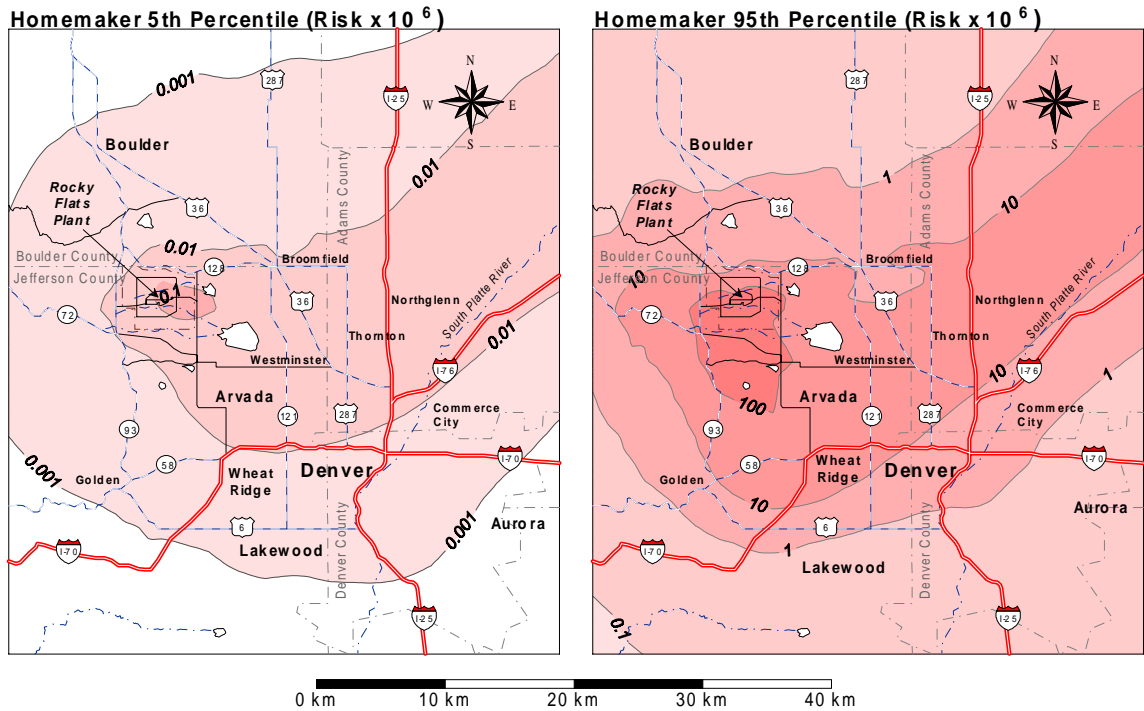
- There is a 90% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was between  $2.2 \times 10^{-8}$  (5% value) and  $2.0 \times 10^{-4}$  (95% value)
- There is a 5% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was greater than  $2.0 \times 10^{-4}$
- There is also a 5% probability the model estimated risk was less than  $2.2 \times 10^{-8}$ .

We may also interpret this to mean, given an exposure history and lifestyle similar to that of the laborer scenario residing in Leyden, there is a 95% probability that the model-estimated number of cancer cases attributed to inhalation of plutonium originating from the RFP would be no greater than 200 persons in a population of 1 million similarly exposed individuals.

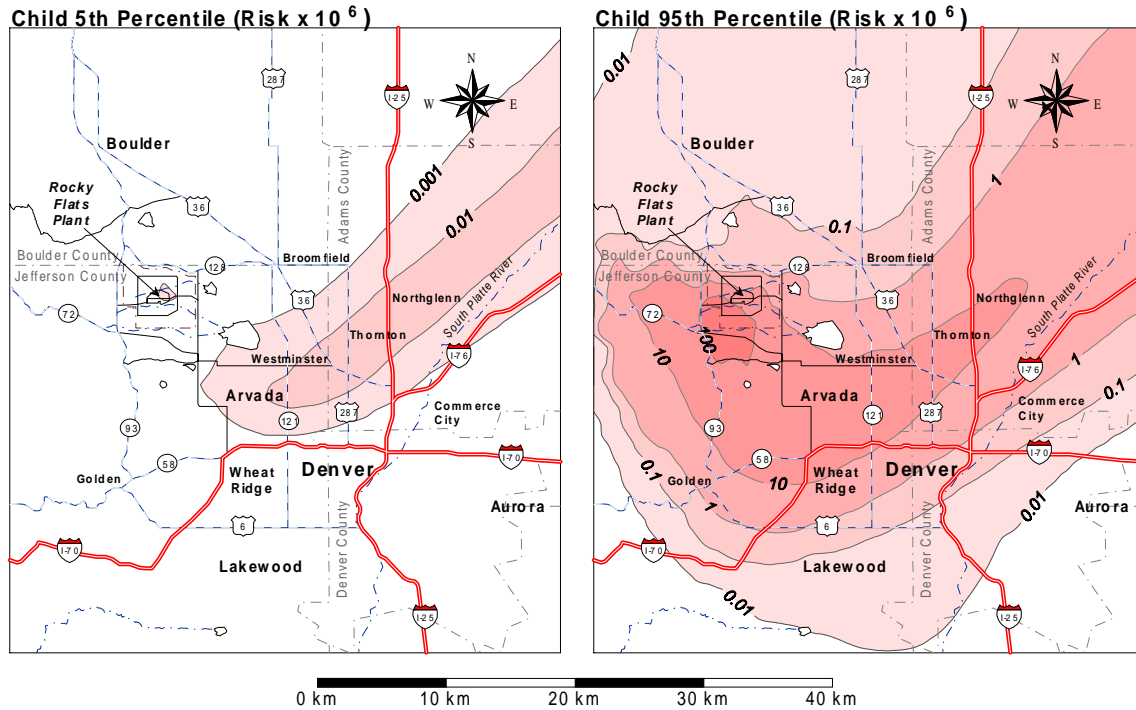
Estimated cancer risks at the 95th percentile level are within the point of departure for acceptable risks established by the EPA of  $10^{-6}$  to  $10^{-4}$ . However, a single grid node near the southwest corner of the RFP boundary had a 95th percentile cancer risk value of  $1.1 \times 10^{-3}$ . The spatial extent of this excursion above the EPA's acceptable risk range was limited to an area no greater than 1 km<sup>2</sup>.



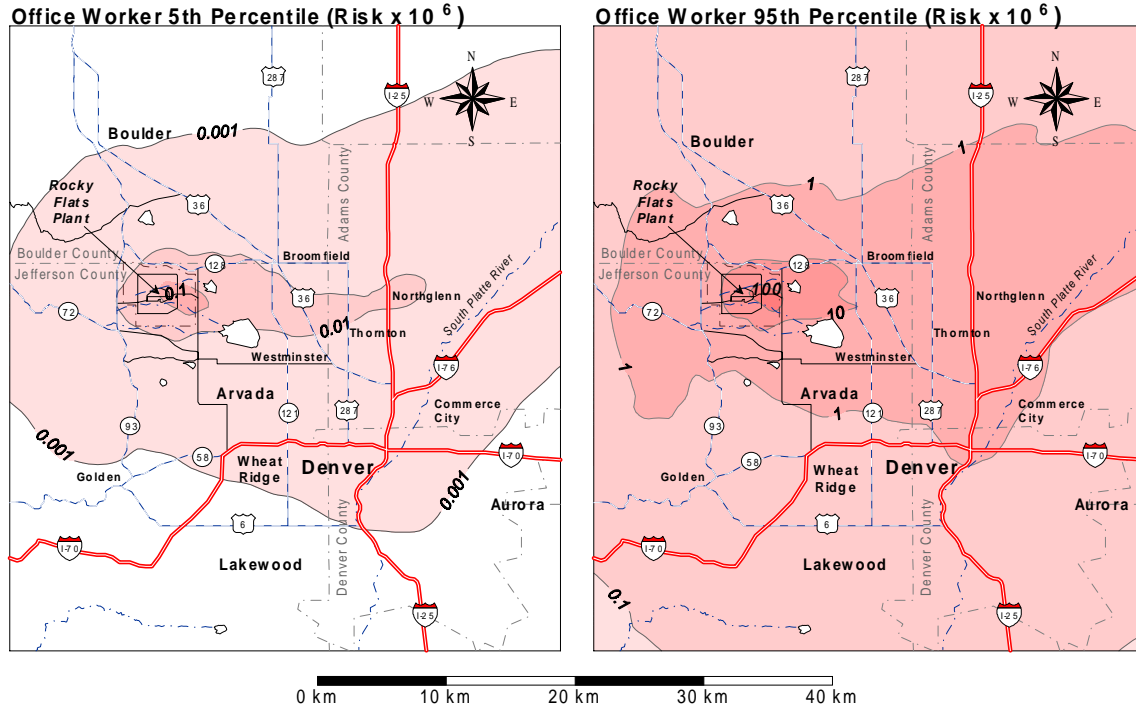
**Figure 28.** Incremental lifetime cancer incidence risk for the laborer scenario; 5th percentile (left plot) and 95th percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 in 1 million chance of developing cancer.



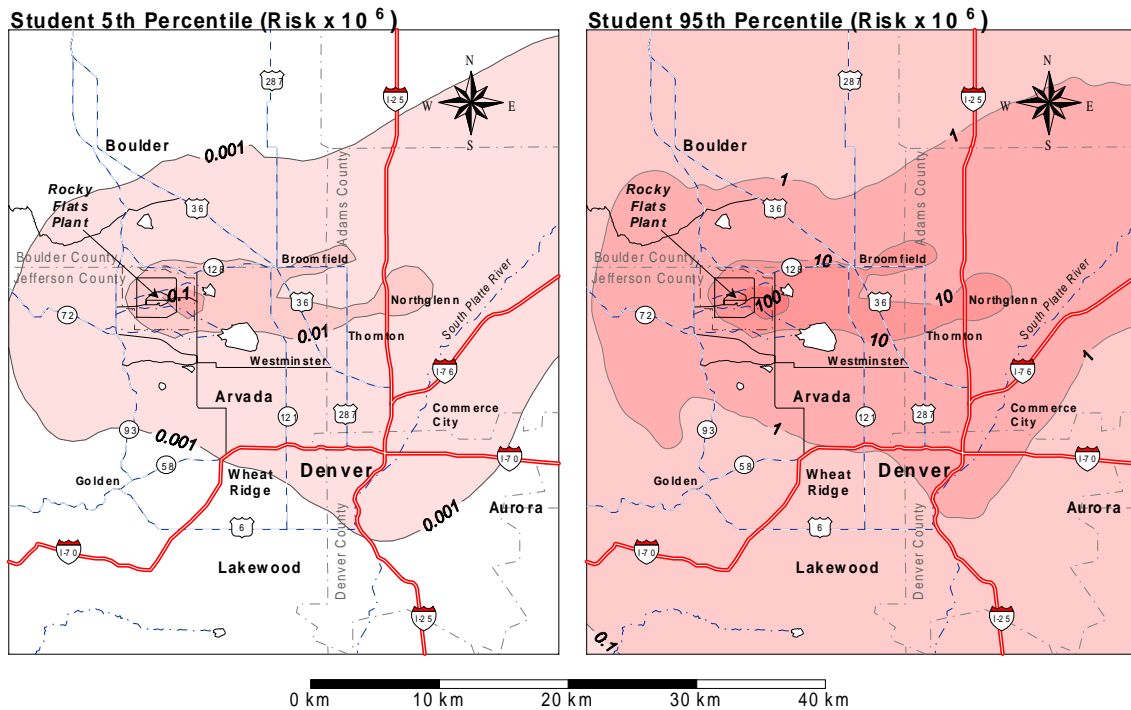
**Figure 29.** Incremental lifetime cancer incidence risk for the homemaker scenario; 5th percentile (left plot) and 95th percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 in 1 million chance of developing cancer .



**Figure 30.** Incremental lifetime cancer incidence risk for the child scenario; 5th percentile (left plot) and 95th percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 in 1 million chance of developing cancer.

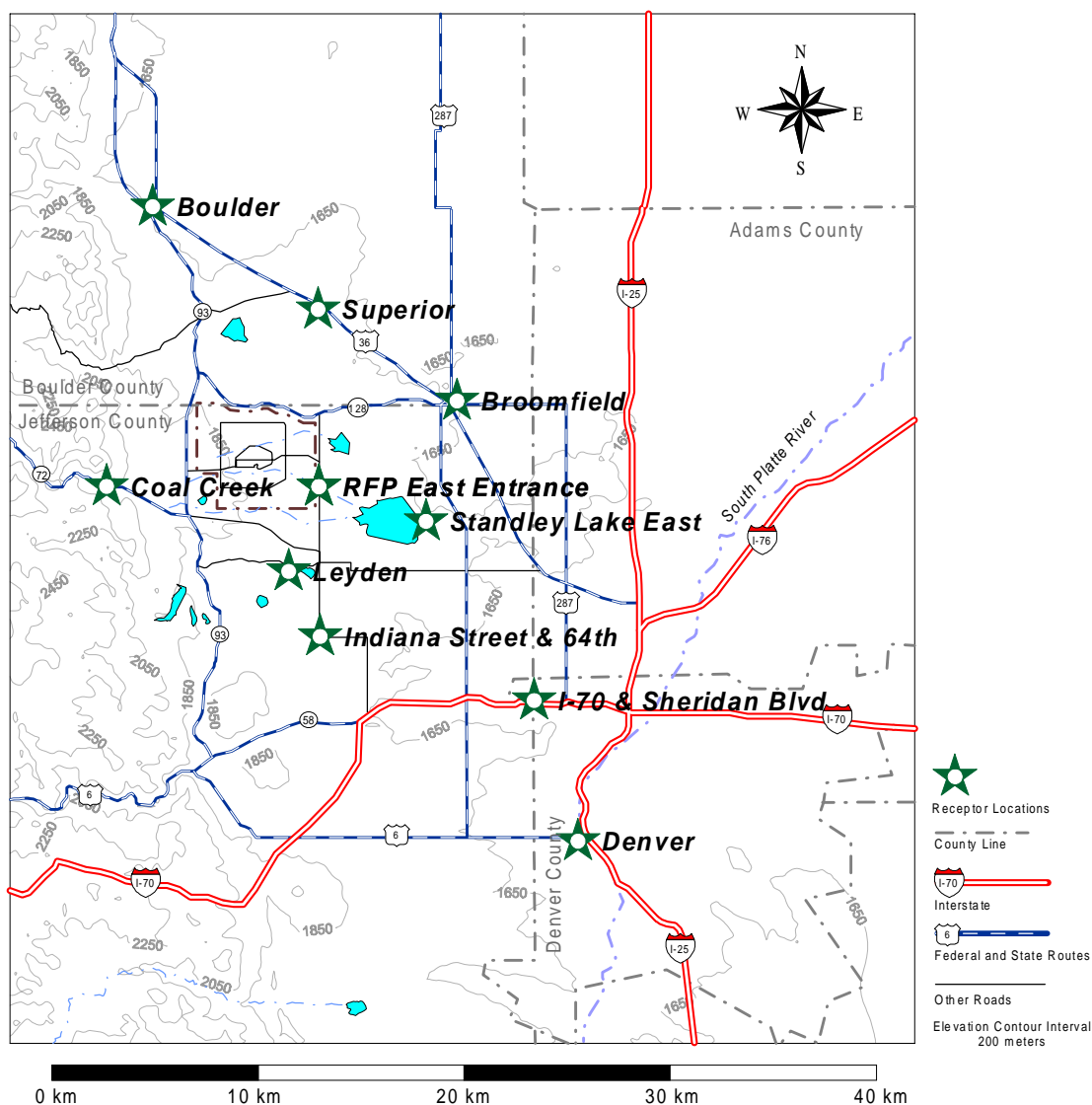


**Figure 31.** Incremental lifetime cancer incidence risk for the office worker; 5th percentile (left plot) and 95th percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 in 1 million chance of developing cancer.



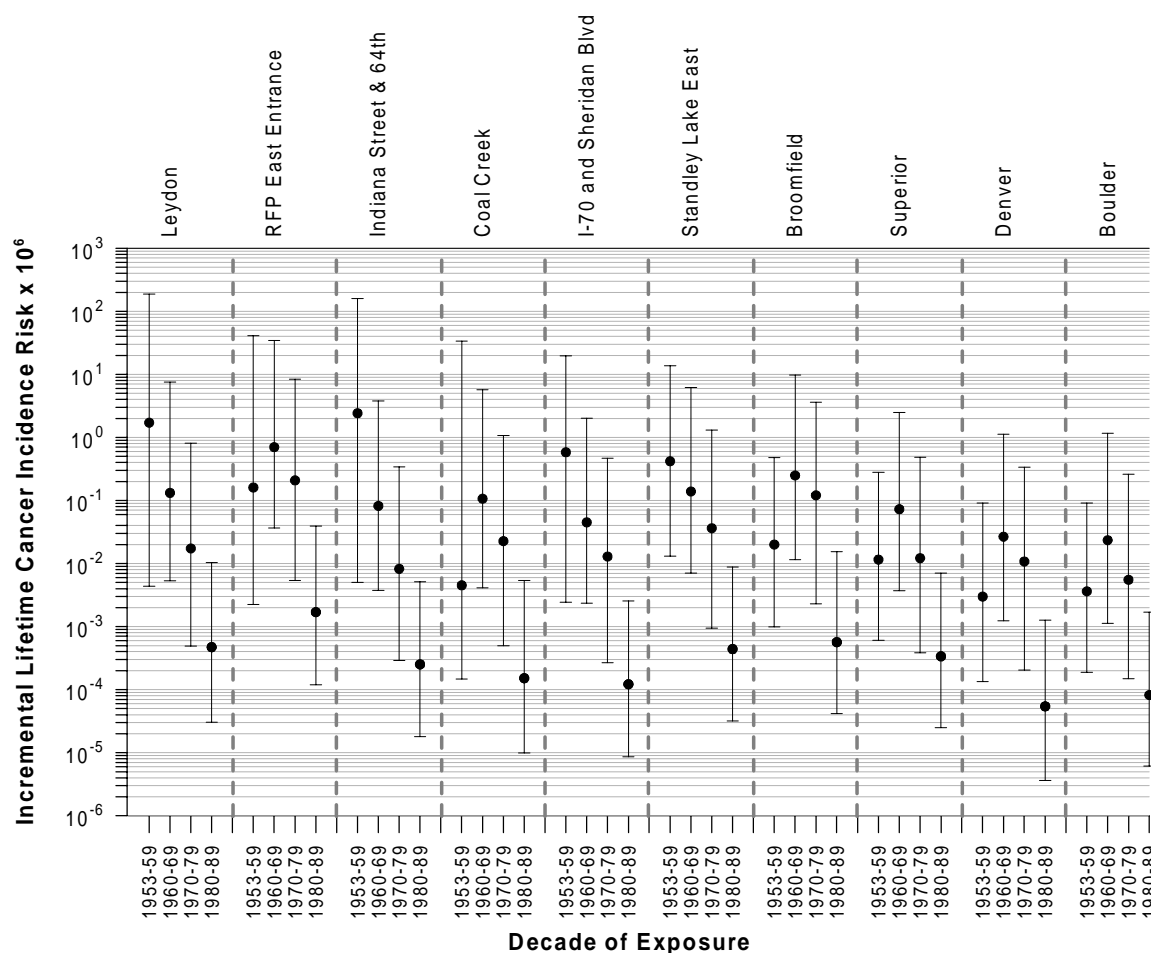
**Figure 32.** Incremental lifetime cancer incidence risk for the student scenario; 5th percentile (left plot) and 95th percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 in 1 million chance of developing cancer.





**Figure 33.** Map showing locations of receptors for evaluating cancer risk as a function of decade of exposure.





**Figure 34.** Incremental lifetime cancer incidence risk for the laborer at selected receptor locations in the model domain. Dots represent the 50th percentile value; vertical bars represent the 5th and 95th percentiles. Cancer risks have been sorted by decade of exposure.

**Table 21. Incremental Lifetime Cancer Incidence Risk for the Laborer Scenario at Selected Receptor Locations in the Model Domain**

Receptor location	Incremental lifetime cancer incidence risk $\times 10^6$ <sup>a</sup>		
	5th percentile	50th percentile	95th percentile
RFP East Entrance	0.097	1.9	91
Superior	0.0069	0.11	3.2
Broomfield	0.021	0.46	20
Stanley Lake East	0.045	0.77	27
I-70 and Sheridan Blvd	0.019	0.72	24
Leyden	0.022	2.3	200
Denver	0.0026	0.05	1.6
Boulder	0.0023	0.042	1.3
Coal Creek	0.01	0.2	59
Indiana Street and 64th	0.026	2.6	150

<sup>a</sup> Risk values have been multiplied by  $1 \times 10^6$ .

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